
P O L I M E R Y

Progress in the development of acrylic resin-based powder coatings – an overview

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Abstract: A review of powder coatings based on acrylic resins and the relationship between the chemical structure of monomers and the structure, properties and use of resins for the production of powder coatings was presented.

Keywords: polyacrylate resins, powder coatings, properties, applications.

Postęp w rozwoju powłok proszkowych na bazie żywic akrylowych – przegląd literatury

Streszczenie: Dokonano przeglądu powłok proszkowych na bazie żywic akrylowych oraz zależności pomiędzy budową chemiczną monomerów, a strukturą, właściwościami i zastosowaniem żywic do wytwarzania powłok proszkowych.

Słowa kluczowe: żywice poliakrylowe, powłoki proszkowe, właściwości, zastosowania.

Coating products are classified according to their form. Coating materials can be distinguished as solvent-based, high-solids, waterborne, and powder coatings. The key role of any coating material is to provide aesthetic or decorative value, offer protection from environmental weathering, and achieve specific performance properties [1]. Coatings should possess good physical and mechanical properties, such as durability, excellent adhesion, weather, chemical and thermal resistance, etc.

However, these are not the only important aspects. After heavy use of petroleum-based synthetic raw materials around the World War II era, health and environmental problems has begun to arise. Earlier, coating materials containing high amounts of volatile organic compounds (VOCs) were used, leading to high emissions and harmful effects on safety, health, and the environment. As a result, several measures have been taken to limit the use of toxic materials such as toluene, hexane, and methanol. Powder coatings, which were first produced in the 1940s as thermoplastic powder coatings and later in the 1950s as thermosetting epoxy powder coatings in Shell's Delft laboratories, meet these requirements perfectly [2]. Powder systems, due to the high-boiling solid compounds (with a boiling point above 250°C), do not emit VOCs [3]. Further, there is no need for biocides, which are otherwise used in liquid systems for protection against the growth of microorganisms. Biocides in liquid coatings can be washed away by water, leading to

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Fig. 1. The characteristics of powder coatings

environmental and water pollution [4]. Powder coatings comply with current regulations, directives, and technical requirements related to specific applications [5, 6].

Furthermore, powder coatings fulfil the “5E” requirements (Fig. 1) [7]:

- efficiency – powder coatings are applied electrostatically, which results in a more efficient and controlled process. The powder adheres to the surface evenly, reducing the need for additional touch-ups or rework, leading to improved productivity and reduced costs.

- economy – although the initial investment in powder coating equipment might be higher compared to liquid coating systems, powder coatings prove to be cost-effective overall. Reduced material wastage, lower energy consumption during curing, and the ability to recycle excess powder contribute to overall cost savings.

- energy savings – curing powder coatings require much less energy as there is no solvent removal involved. Powder coatings do not require drying time like common liquid systems, reducing production time.

- environmental compliance – one of the most significant advantages of powder coatings is their eco-friendliness. Unlike liquid coatings that contain VOCs, powder coatings are solvent-free and emit negligible amounts of VOCs. This makes them a more sustainable and environmentally responsible option, reducing air pollution and minimizing health risks for humans and the environment.

- excellence of finish – powder coatings exhibit good properties, particularly better adhesion and corrosion protection compared to liquid coatings. Once cured, they form a tough and resilient film that can withstand harsh environmental conditions, including exposure to UV radiation, moisture, chemicals, and extreme temperatures.

The increasing demand for environmentally friendly and durable coating solutions, coupled with the growth in industrial and construction activities, has contributed to the rise in the powder coatings market. Currently, powder coatings play key role of the chemical industry. The total value of the global coatings market was estimated to be \$160 billion in 2021 [8]. In 2020, the global powder coatings market size was valued at \$11.9 billion. The annual growth rate (CAGR) in the production of powder coatings products on a global scale is expected of 7.2%. According to market predictions, production is expected to reach \$20.8 billion in 2028 (Fig.2) [9]. In 2022, the North American and Latin American powder markets showed growth of 3.5% and 1.8%, respectively [10]. As industries and consumers continue to prioritize sustainability and long-term

cost-effectiveness, the popularity of powder coatings is expected to continue grow globally.

The increase in production of powder coatings can be attributed not only to the advantages offered by the “5E” characteristics, but also to the continuous development and innovation in the field. As companies and scientists focus on improving powder coating technologies, they unlock new possibilities and expand the applications of powder coatings in various industries. Recent developments in powder coatings include:

- low-temperature cure – technologies like Crylcoat produced by Allnex allow for curing at lower temperatures, which can be advantageous for temperature-sensitive substrates or to reduce energy consumption during the curing process.

- UV cure technology – UVECOAT produced by Allnex utilizes UV light for curing, enabling faster curing times and potential applications in industries where quick turnaround is essential.

- primers with anti-corrosion system – products like Alesta® Zero Zin produced by Axalta offer enhanced corrosion protection and serve as effective primers for demanding and aggressive environments.

- recycling and sustainability – the development of powder coatings with recycled content, such as Powdura®ECO produced by Sherwin-Williams, contributes to sustainability efforts, and supports the circular economy by utilizing post-consumer recycled materials [10].

Powder coatings are used in a wide range of industries, including automotive, furniture, electronics, industrial and household equipment, agricultural equipment, etc. Their versatility and ability to meet specific performance requirements make them an attractive choice for various applications [11]. As technology continues to advance and more research is conducted, further innovations and improvements in powder coatings is expected, making them an increasingly popular and sustainable choice for surface finishing across multiple industries.

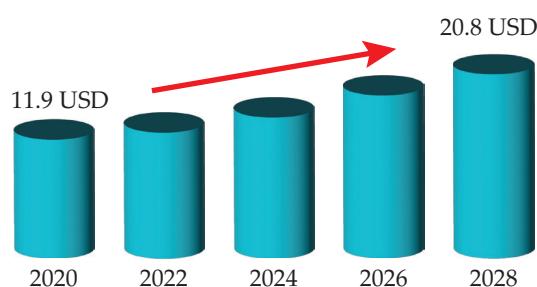




Fig. 2. The powder coatings market forecast for 2020–2028 [9, 10]

Table 1. Standard thermosetting powder coatings [15]

Resin	Functional group of resin	Hardener	Advantages	Disadvantages
Polyester	-COOH	TGIC Promid	Volatile free, Mechanical property	Toxicity, Bubbling
	-OH	PIC blocked NCO Aminoplast	Smooth appearance Mechanical property	Volatiles Storage stability
Epoxy		Dicyandiamide anhydride	Corrosion resistance Volatile free	Exterior durability Yellowing
Acrylic	-OH	PIC blocked NCO Primid	Exterior durability Exterior durability	Impact resistance
	-COOH			Mechanical properties
		UV-curing photoinitiator	Exterior durability Corrosion resistance Good adhesion	Mechanical properties

Powder coatings are defined as a mixture of 100% solid components [11]. Thermosetting powder coatings are formulated using resins that contain functional moieties that react with other groups derived from the cross-linker agent. Thermoplastic powder coating is made of thermoplastic resin, pigment, filler, plasticizer, and stabilizer [12]. Thermosetting powder coatings are most often used because of their much better physical and mechanical properties than thermoplastic systems [13]. UV curing is also becoming increasingly popular [14].

There are three main types of resins in thermosetting powder coatings: epoxy, polyester and acrylic (Table 1). Among them, polyester resin is used more often than others due to the cost/performance relationship. The hydroxy-functional polyester resin commonly cross-linked by blocked aliphatic isocyanates exhibit excellent mechanical properties and abrasion resistance, which are typical of polyurethane coatings [16]. Recently, the most widely used cross-linker for carboxylic acid-terminated polyesters was triglycidylisocyanurate (TGIC). TGIC-based powder coatings have good exterior durability and mechanical properties [17]. Nevertheless, a powder coating company in Europe has withdrawn the production of TGIC, deeming it as a toxic hazard. An example of polyester resins for powder coatings without TGIC is the Interpol Powder Coatings [18]. Epoxy powder coatings are characterized by very good mechanical properties, adhesion, and corrosion protection, but yellowing under UV radiation is a challenge [19, 20]. For this reason, epoxides are better suited for indoor applications [21].

Acrylic resins, when compared to other powder systems based on epoxy or polyester resins, have functional groups distributed along the length of the chain, which allows for higher cross-linking density [16]. Acrylic powder coatings characterized by superior weatherability, chemical, thermal, and mechanical resistance. It is proved by the hydrophobic properties, color durability and high resistance to scratches, sand blows and damage caused by stones. These

coatings find applications in the architectural, automotive industry and heat-sensitive substrates such as wood, plastics, and medium-density fiberboard (MDF) [22]. Acrylic resins for powder coatings can be divided into three types: hydroxyl group-containing, carboxyl group-containing, and glycidyl group-containing acrylic resins. Depending on the specific functional group used, polyacrylate for powder paints can exhibit different properties, which influences their application [23].

This work aims to investigate the impact of selection of acrylic monomers on the formulation of acrylic resin for powder coatings, with the objective of achieving enhanced physical and mechanical properties. By understanding the relationship between acrylic monomers, resin formulation, and polymerization methods, the review seeks to optimize the production process and develop powder coatings with superior properties, such as improved weather resistance, chemical resistance, color durability, hydrophobicity, and mechanical strength. The findings of this study can have significant implications for industries such as architecture and automotive, and for the coatings for heat-sensitive substrates like wood, plastics, and medium-density fiberboard (MDF).

CHOICE OF MONOMERS

The substantial number of monomers makes it possible to prepare acrylic resins with unlimited possibilities. For these reasons, manufacturers can design resins to meet the specific requirements of the final product. To fully harness the potential of acrylic resins, a profound understanding of the chemical structure of their constituent monomers is paramount. This knowledge becomes the key to unravelling the intricate correlation between the resin's properties and its ideal applications. The acrylic monomer contains vinyl groups, which directly connecting with to the carbonyl carbon of the ester group. This unique arrangement imbues acrylic monomers with

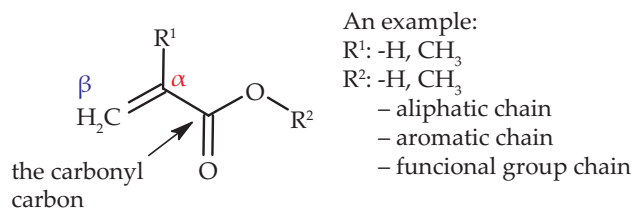


Fig. 3. Chemical structures of methyl/acrylate monomers

a bifunctional nature, where the vinyl group effortlessly facilitates free radical polymerization, expediting the formation of the acrylic resin. Simultaneously, the carbonyl group unlocks a myriad of functionalities, offering a canvas for creative transformations through interactions with amines, acids, or alcohols (Fig. 3) [24].

Moreover, the sidechains on monomers in acrylic resins plays a pivotal role in determining various properties of the resulting polymers. Acrylic resins that contain monomers with short sidechains tend to form polar polymers. On the other hand, acrylic polymers composed of methacrylic derivatives are even more polar than those containing acrylic derivatives. This polarity affects several aspects of the acrylic resin's behavior, including its solubility and interaction with other materials. Acrylic polymers containing long aliphatic sidechains exhibit a distinct nonpolar nature. Consequently, they display a greater affinity for nonpolar solvents, leading to their enhanced solubility in such environments. Another significant effect of long aliphatic sidechains in acrylic polymers is the notable reduction in viscosity, both in organic solutions and in melts. This reduced viscosity simplifies the processing and handling of acrylic resins during manufacturing and fabrication [25].

Esters of acrylic acid

Butyl Acrylate (BA) stands as one of the principal acrylic monomers utilized in powder coatings [26]. Its inclusion in acrylic resin aims to enhance the impact strength of acrylic materials [27]. The flexibility properties of polymers containing BA are significantly influenced by the chain length and branching of alcohol sidechains. A study conducted by Li and co-workers delved into the effects of alkyl chain length in acrylate monomers [28]. In this research, various acrylate monomers were grafted with alkyl chains of different lengths (e.g., methyl acrylate (MA), ethyl acrylate (EA), butyl acrylate (BA), and ethylhexyl acrylate (EHA) onto native feather keratin. The results showed that as the alkyl chain length increased, the tensile strength, tensile elongation, and abrasion resistance of the polymers improved. Furthermore, an increase in the ester side chain length of acrylic monomers, [2-(phenylcarbamoyloxy) isopropyl acrylate (PhCPA) and 2-(hexylcarbamoyloxy)isopropyl acrylate (HCPA)], was correlated to an increase in the propagation rate coefficient during polymerization [29]. Azeri *et al.* also investigated the effect of acrylate chain

length using butyl and dodecyl acrylate, focusing on the hydrophobicity effect [30]. They discovered that longer acrylic monomer chains led to greater repulsion of polar particles.

Esters of methacrylic acid

Poly(methyl methacrylate) (PMMA) stands as a typical example of a polymer based on methacrylic acid ester. In many cases, it is co-polymerized with methyl methacrylate (MMA) to achieve the desired film hardness. PMMA finds extensive application in various industries, including biomedicine (dental fillings, optically clear sheeting, limb prostheses), electronics (computer-to-sensor data transmission links), bathroom fittings (bathroom sinks and shower surrounds), and others [31].

It is worth noting that all esters of methacrylic acid polymerize at a slower rate compared to acrylic acid esters. This effect can be attributed to steric hindrance caused by the presence of a methyl group in the α -vinyl position of the monomer. Kostakis *et al.* investigated the reactivity ratios of methyl methacrylate with cyclohexyl and trimethylsilyloxy ethyl methacrylate using various graphical methods such as Finemann–Ross, inverted Finemann–Ross, and Kelen–Tüdös [32]. The steric hindrance induced by the cyclohexyl ester group and other bulky side groups reduces the polymerization rate and can even impede the reaction altogether. This phenomenon is also observed in other bulky monomers like *t*-butyl methacrylate, trimethylsilyl methacrylate, and stearyl methacrylate (SMA) [33]. Nevertheless, the use of five-chain monomer methacrylate increased the tensile strength (14.31 MPa) similarly to acrylates monomers, as described by Bi *et al.* [34].

Functional monomers

The functional monomers play a key role in the curing reaction of powder coatings [35]. The composition and functionality of these monomers in the formulation significantly influence the degree of crosslinking in the resulting coating structure [36]. Increasing the functionality of the monomers leads to higher crosslinking density, meaning there are more conjunction points within the polymer network. Additionally, this results in an increase in the glass transition temperature (T_g) of the coating as the mobility of the polymer chains are reduced. The higher crosslinking density also have a positive impact on properties such as hardness, scratch resistance, and chemical resistance, making the coating more durable and robust [37]. Johanson *et al.* studied the effect of acrylate functional hyperbranched polyesters on the end-groups: hydroxyl, propionate, or benzoate groups, and found that it can be utilized to modify cross-linking density and polarities of the films. Depending on the structure of the non-reactive end-groups, T_g of the final films are between 67 and 77°C. Changing the end-groups from hydroxyl groups to propionate only shifts

the T_g 10°C. The major factor affecting the T_g was type and amount of cross-linking [38].

Hydroxyl-functional monomers

In the 1960s, many copolymers made from acrylic/methacrylic monomers containing hydroxyl groups were developed and became the most important class of resins, known as solution polymerization products [27]. Initially, melamine resins were the most important crosslinkers for hydroxy-functional acrylic resins due to their excellent weatherability, which complemented the properties of acrylic resins [39].

Currently, acrylic resins containing free hydroxyl groups are crosslinked with blocked polyisocyanates, and sometimes amine compounds are also used in powder coatings [40, 41]. The hydroxy-functional monomers differ in their structure and the reactivity conferred by the type of hydroxyl group. These differences play a crucial role in their applications in the production of acrylic resins. When comparing hydroxy-acrylic resins with comparable viscosities and the same hardener, their pot-lives increase in the following order: 4-hydroxybutyl acrylate < 2-hydroxyethyl acrylate < 2-hydroxyethyl methacrylate < 2-hydroxypropyl acrylate < 2-hydroxypropyl methacrylate. The high reactivity of 4-hydroxybutyl acrylate is due to the exposed primary hydroxyl group. While high reactivity is advantageous, it can also lead to shorter pot-life, which can be disadvantageous for application properties, not only due to time restrictions, but also in terms of achieving optimum film formation, flow, and levelling [27, 42].

Carboxy-functional monomers

The production of carboxyl-functional acrylic monomers saw significant growth in the late 1950s, primarily driven by the demand for superabsorbent. Acrylic acid and methacrylic acid began to be produced on a global scale to meet this purpose [43]. In recent literature, superabsorbent polymers have garnered attention due to their unique properties, such as their ability to absorb and retain substantial amounts of water along with high volume expansion. These superabsorbents have been utilized as additives for the repeated barrier restoration of damaged powder coatings under wet-dry cycles. This approach shows promise for potential applications in external healing concepts, particularly in the case of scratches and corrosion reactions. [44].

Like the hydroxyl groups, carboxyl groups also facilitate crosslinking reactions with appropriate curing agents. In general, when we replace the hydrogen in the -OH group by a carboxyl group, an O-C bond is formed in place of the O-H bond [45]. For this reason, there are many polyester resins on the powder paint market (e.g., Siraless), although not always based on acrylates or methacrylate's, due to their cost factor [46].

Epoxy-functional monomers

The most common methacrylic monomer used in coatings is glycidyl methacrylate (GMA). The epoxy-functional monomer is industrially produced and impart good mechanical properties such as rigidity, toughness, and adhesion to acrylic resins. The presence of polar hydroxyl and ether groups in the backbone structure of epoxy resins contributes to their excellent adhesion performance. As a result, acrylic resins containing glycidyl groups find potential applications in waterproof and anti-corrosion coatings [47]. However, when we add, for example, multi-walled carbon nanotubes to acrylic resin – glycidyl methacrylate (GMA), it improves the electrical, mechanical, thermal, and hydrophobic properties of coatings [48].

Another critical aspect that significantly influences the properties of coatings is the choice of curing agent. The nucleophilic ring opening reactions of the epoxide group in glycidyl methacrylate (GMA) allow for various modifications with different functional groups, such as amine-epoxy, thiol-epoxy, azide-epoxy, acid-epoxy, and hydrolysis reactions [49]. The versatility of modification and curing reactions with GMA enables coatings to be cured using low/high temperature and UV curing technologies [50, 53]. The use of an ionic photoinitiator in the reaction of the epoxy group during cationic polymerization can overcome problems associated with oxygen inhibition compared to free-radical photopolymerization.

A study conducted by Bednarczyk *et al.* involved the synthesis of epoxy methacrylate resin and coatings preparation using both cationic and radical photo cross-linking. The research demonstrated that while cationic polymerization was slower with a lower conversion of photoreactive groups compared to radical polymerization, the choice of polymerization type did not really influence the final properties of the coatings. All the coatings exhibited good properties [51].

Special monomers

Special monomers like 2,2,2-trifluoroethyl methacrylate (FMA) or siloxane (meth)acrylic introduce unique surface properties to coatings [52, 53]. Fluoropolymers, such as those derived from FMA, possess exceptional characteristics like high thermal, chemical, aging, and weather resistance, low dielectric constant, refractive index, surface energy, and flammability [54]. Silicone-containing coatings characterize by efficient water vapor permeability and self-cleaning properties [55].

A study by Yu *et al.* explored the influence of silane components, specifically γ -methacryloxypropyl triisopropoxidesilane (KH571) with itaconic acid (IA), on the properties of waterborne acrylic resin. The investigation included assessing mechanical properties, water resistance, thermal stability, storage stability, and corrosion resistance.

The contact angle of the resin coating increased from 78.91 to 90.49°, indicating enhanced hydrophobicity [56]. In

Table 2. Polyacrylate resins and their properties

Type of resin	Example of polymer	Properties	References
Based on esters of acrylic acid	The modified feather keratin-g-poly(AA-coacrylate) grafted with acrylates with different alkyl chain lengths	High the tensile strength, elongation, and abrasion resistance	[29]
	The amphiphilic block copolymers of the type of poly (alkyl acrylate)-b-poly (acrylic acid) (PAlkA-b-PAA) and (acrylic acid) (AA)	Increased the hydrophobicity	[31]
Based on esters of methacrylic acid	poly(methyl methacrylate) (PMMA)	The high scratch and chemical resistance, hardness, and good thermal stability	[32]
Based on hydroxyl-functional monomers	Polyacrylate resin based on 2-hydroxyethyl methacrylate (HEMA) Curing agent: blocked polyisocyanates	The excellent weather durability, better flexibility than polyacrylate contain carboxy- or epoxy-groups	[41]
Based on carboxy-functional monomers	The polymers based on acrylamide/acrylic acid	The good transparency and chemical resistance, potential applications in self-healing concepts	[45]
Based on epoxy-functional monomers	The acrylic resin modified with glycidyl methacrylate (GMA)	The good corrosion protection, weather durability and stain resistance	[48]
Based on special monomers contained fluorine atoms or silica atoms	Fluoropolymers based on 2,2,2-trifluoroethyl methacrylate (FMA)	The high thermal, chemical, aging, and weather resistance	[55]
	The polymers based on γ -methacryloxypropyl triisopropoxidesilane (KH571) and itaconic acid (IA),	The good mechanical properties, water resistance, thermal stability, storage stability, and corrosion resistance	[57]

another study by Zhou *et al.*, acrylic-based polyurethane coatings with nano-silica particles were prepared. The addition of nano-silica led to apparent improvements in macro-hardness, micro-hardness, abrasion resistance, scratch resistance, tensile strength, and Young's modulus [57].

However, too much silica/fluorine or various additives used in powder coatings can have counter-productive effect too. The introduction of a fluorine or silicone atom as a heteroatom into the hydrocarbon chain can lead to incompatibilities [56]. Xie *et al.* reported a solution to this problem. In their work, they found that encapsulating additives with the same resin of the host powder coating can enhance both flowability and film properties, compared to commercial nano-silica [58].

Selected properties of polyacrylic resins

Table 2 summarizes the differences resulting from the structure of monomers used for given types of polyacrylate resins and their selected properties. This summary was created to better monitor the development in this field. In the case of polyacrylate resins with functional groups (-OH, -COOH and epoxy group) a cross-linking agent also affects the final properties. The effect of cross-linking agents between polyacrylate resins containing functional groups are described in depth later in this review article.

POLYACRYLATE RESINS

According to Poth, acrylic resins for coatings are polymers composed of esters of acrylic acid or methacrylic acid. Depend on used esters of acrylic acid and meth-

acrylic acid are distinguished by the reactivity of its double bonds. The reactivity increases as follows: acrylic > methacrylic > vinyl >> allyl [27]. The higher the reactivity of the monomers, the quicker the polymerization process occur. For this reason, the condition (e.g., initiator concentration, polymerization temperature, inert atmospheres etc.) of polymerization is truly relevant [59]. Uncontrolled reactions can cause undesirable effects. Moreover, acrylic monomers and comonomers can polymerize spontaneously under moisture, susceptible to oxygen and temperature influences. Therefore, the synthesis of polyacrylates is most often carried out in an atmosphere of an inert gas such as nitrogen. Monomers, on the other hand, are stabilized by hydroquinone derivatives, due to their wider solubility [60]. Acrylic resins have distinguished two synthesis methods. The first method of acrylic resins is prepared by polymerizing acrylic or methacrylic esters via their double bonds. The second group of acrylic resins for coatings still contain unreacted/free double bonds [27].

Methods of synthesizing acrylic resins by polymerization of double bonds

The chain growth polymerization is the most used chain reaction for making vinyl copolymers (often acrylics) and containing three types of chemical reactions – initiation, propagation, and termination (Fig. 4) [61, 62]. The first step, initiator (peroxy- or azo- compounds) reacts to form an initiating free radical. Free radical reacts rapidly with the π -electrons of the double bonds of monomers to form a second free radical. The double-bond in acrylic creates a tetrahedral molecule of lower bond energy. Thus, the amount of energy is released in the form of heat (exo-

thermic reaction). Propagation reactions are fast because acrylic monomers are very reactive. The chain grows until forming a single σ -bond. The final stage is termination of growing chain by recombination and disproportionation. Only in the case of solution polymerization, hydrogen transfer based termination is possible. Disproportionation is a reaction within two neighboring molecules, wherein one free radical becomes a saturated chain end.

The polymerization reaction depends on numerous factors, e.g., initiator concentration, polymerization temperature and the environment in which the reaction is conducted. The presence of a solvent has a profound influence on the course of polymerization. The literature showed that as the length of the ester chain increases, the propagation rate coefficients in the bulk increases or is similar that in the toluene solution [63, 64]. As a result, for short ester chains (up to C4) the propagation rate coefficient is higher in solution than in bulk polymerization. Exceptions are acrylates with long chains (over C16) and at lower temperatures [66]. In this case, there is evidence to suggest that the propagation rate coefficient in bulk and solution may be similar. In addition, the propagation step of acrylates in the solvent can be affected by hydrogen bonding, which increases the propagation rate coefficients [65].

Using solvent polymerization for powder coatings, it is necessary to remove the solvent by e.g., by using distillation [66]. Nevertheless, solution polymerization can result in polymers with small molecular weights compared with those of bulk polymerization or suspension polymerization. The selection of the right solvent is also critical. Aliphatic hydrocarbons and terpene hydrocarbons, the residual solvent classes, are unsuitable for acrylic resins [67]. The better option are alkyl aromatic solvents, but compounds such as xylene, cumene, trimethylbenzene or methyl iso-butyl ketone are harmful for people [68]. Thus, they are restricted in use to comply with VOC regulations. In recent years, there has been an increase in the produc-

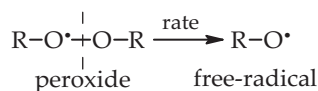
tion of water-based acrylic resins, which are not as harmful e.g., SETAQUA® 6799 by Allnex company [69].

Polymerization in the bulk of methacrylic or acrylic monomers is characterized by high degree of conversion [70]. The initiator is added to the meth/acrylic monomers without solvent, the viscosity and molecular mass increases. The disadvantage of this method is the problem with heat dissipation, causes e.g., bubble formation in acrylic resin [71]. Nevertheless, the use of bulk polymerization in powder coatings gives many advantages: no evaporation of solvent required (environmental aspect, energy efficiency) and high molecular weight (desirable feature in manufacture of powder coatings). The effect of the decreasing of free monomer content, which yields a lower propagation rate, is compensated by a rise in viscosity, resulting in a lower termination reaction velocity. The most common use for bulk polymerization is manufacturing the so-called acrylic glass, consisting of poly methyl methacrylate [72]. This polymerization process is most preferred for solvent-free secondary dispersions or powder coatings. To cite an example would be the company's Color & Comfort product range: 1) glycidyl methacrylate (GMA) functional acrylic resins for curing with dicarboxylic acids FINEDIC FINEPLUS, 2) acrylic powder resins compatible with FINEDIC FINEPLUS polyester powder coatings) [73].

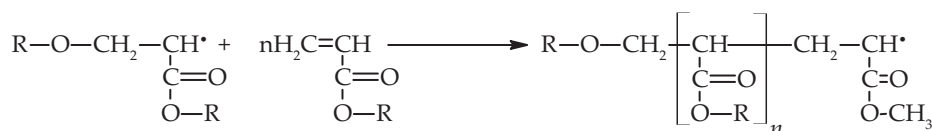
Methods for the synthesis of acrylic resins with unreacted/free double bonds

The second group of acrylic resins for coatings still contain unreacted or free double bonds. Copolymerization of polyacrylate utilizes the addition or condensation reaction mechanism. Multifunctional monomers find use in medicine applications in medicine, pharmaceuticals, paints, column packing, optics, polymer additives or ion exchange resins [74]. Free-radical initiated polymerization process occurs in seconds, but polycondensation

Initiator reaction



Propagation



Termination

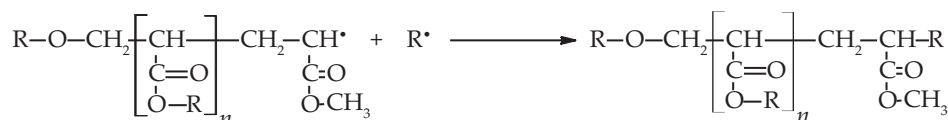


Fig. 4. Scheme of polymerization steps [65]

reactions take hours. The choice of type of copolymerization depends on the functional group of monomers. Comparing the course of radical polymerization and polycondensation, the monomer concentration decreases gradually during the radical polymerization process and the polymer yield increases with increase in reaction time. The polycondensation yield is independent of the reaction time. The conversion of functional groups reacted during polycondensation is compared to their initial amount. Polyaddition, like polycondensation, is a step-by-step process. The molecular weight of the polymer increases evenly. Moreover, free-radical initiated polymerization is not equilibrium process, in contrast to polycondensation [75].

The polymerization is initiated by energy-rich radiation (e.g., UV light), which yields three-dimensional crosslinked macromolecules. The commercial example of a (meth)acrylate polyester powder coating resin is UVECOAT 2100, supplied by Allnex company. The cross-linking reaction of powder coatings between UVECOAT 2100 and Irgacure 184 or Irgacure 819 under the influence of UV radiation was described by Mehr *et. al* [76].

CHEMICAL STRUCTURE OF ACRYLIC RESIN AND ITS INFLUENCE ON THE FORMULATION AND PROPERTIES OF POWDER COATINGS

According to the definition of powder coatings, they are composed of solid raw materials. The powder coating production process consists of several stages. Initially, the solid raw materials are mixed before being homogenized using an extruder. During extrusion, the temperature of the extruder shaft is raised stepwise to a pertinent temperature (e.g., between 70 and 120°C) to allow the raw materials to mix and melt. The homogenous melt obtained from the extruder is then cooled and solidified.

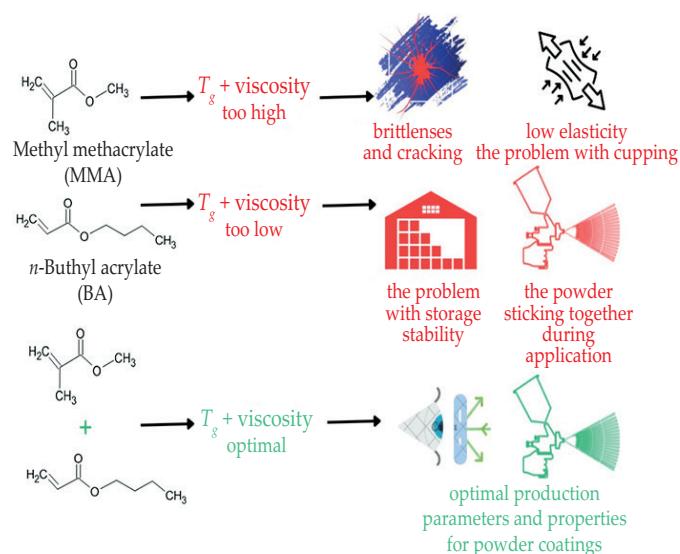


Fig. 5. Chemical structure of appropriate acrylic monomers influences on processing and properties of powder coatings based on polyacrylate resins

The material is subsequently flaked and ground into solid particles of the desired size. The most common method of application for powder coatings is electrostatic spray application. The coated product is then cured in an oven, where the solid powder paint particles transform into a strong and smooth coating [77, 78].

The core component of powder coatings is the resin, which must be processable using suitable methods to produce a stable powder. In thermoplastic systems, the resin melts, flows, and levels during film formation. In thermosetting systems, an appropriate cross-linking agent is used depending on the choice of resin. The appearance and functionality of powder coatings depend on the rheology of the starting material formulations. While additives such as pigments, catalysts, and fillers are also important in powder formulation, the selection of resin (binder) and crosslinking agent play the most crucial role [79].

The parameters, such as the glass transition temperature (T_g) and viscosity of the binder system, determine the storage stability of the powder coating, its ability to adhere during application, and suitable film formation. Any inadequacy in such parameters of the powder coating can lead to defects, such as surface defects caused by incomplete particle sintering or coalescence, as well as 'back ionization' phenomena observed in electrostatic application [80]. This problem not only affects the appearance of the sample, but also its final properties such as abrasion and scratch resistance [81].

The glass transition temperature (T_g) and viscosity are closely related to the molecular weight of the resin. Understanding the correlation between molecular weight distribution, viscosity, and the determination of the glass transition temperature of the resin can provide essential information regarding the formulation and properties of powder coatings, including internal stresses of polymer layers, abrasion resistance, impact strength, and mechanical strength. For this reason, the following subsections will discuss the influence of the chemical structure of acrylic resins and their parameters on the formation of powder coatings. Figure 5 shows, the most used acrylic (nBA) and methacrylic (MMA) monomers, and their chemical structure effect on the T_g and viscosity as well as the impact of these parameters affect the application, storage, and properties of the final product.

Molecular weight distribution

Chromatographic techniques play a vital role in determining the molecular weight distribution and composition of acrylic resins used in powder coatings [82]. Gel permeation chromatography (GPC) is a commonly used method to analyze the molecular weight distribution and dispersion of synthesized resins, enabling researchers to assess the reproducibility of the obtained resins.

The molecular weight of the resin used for powder coatings is in the range of 10,000 and 200,000 Da. It is the most optimal volume, because too low a weight causes

uneven powder coating (poor surface coverage) and on the other hand, high mass form too thick a layer or result in reduced flow during melting [83]. The method of bulk polymerization can yield acrylic resins with high molecular weight distribution values, but it can also lead to highly exothermic reactions, which may cause product reproducibility issues. On the other hand, the solvent polymerization method may result in acrylic resins with low molecular weight, requiring the removal of the solvent before use.

Additionally, the particle size distribution plays a crucial role in the polymerization process. A narrow particle size distribution is advantageous in creating new polymer systems. For example, Tang *et al.* synthesized polyacrylate powder coatings via one-step mini-suspension polymerization, resulting in powder coatings with regular spherical morphology particles and narrow particle size distribution. This narrow distribution positively influenced the fluidity of the powder and the smoothness of the surface layer [84]. By carefully controlling the molecular weight distribution and particle size distribution, researchers can optimize the properties and performance of acrylic resins for powder coatings, ensuring excellent application characteristics and final coating quality.

Viscosity

Another parameter affecting the formulation and storage of powder coatings is the viscosity. The viscosity of powder coatings must be carefully controlled to ensure optimal application and film formation. If the viscosity is too low, the powder particles may stick together, leading to problems with storage and application. On the other hand, if the viscosity is too high, the coating may not melt adequately during the curing process, resulting in difficulties forming a thin, smooth film.

The molecular mass distribution and polydispersity of acrylic resins are closely related to viscosity. Methacrylic polymers have lower chain mobility due to the presence of a methyl group on the sidechain opposite the carboxyl group, resulting in lower polymerization velocity compared to acrylic derivatives. Therefore, the viscosity of an acrylic resin based on methacrylic monomers could be lower than that of a resin based on acrylic monomers, even though the latter has lower molecular weights [85].

In powder coatings, the viscosity cannot be adjusted easily by using organic solvents as in solvent-based paints because powder coatings do not use organic solvents. Instead, other methods must be employed to control viscosity. The viscosity of powder coatings increases with the strength of interactions between chain segments, such as polarity and hydrogen bonding, and with increasing molecular weight [86]. Thus, it can be concluded, that by selecting the appropriate resin, through its chemical structure and the interaction of chain segments, the viscosity of powder coatings can be controlled.

Johanson *et al.* described influences on the chemical structures of an amorphous methacrylate-functional prepolymer and crystalline acrylate and methacrylate monomers on the properties of the powder coatings. The introduction of a crystalline component in an amorphous resin mixture demonstrated reduction of the melt viscosity and enhancement of the flow properties of the powder coatings [87].

Glass transition temperature

The glass transition temperature (T_g) of an acrylic resin for powder coatings is particularly important for many reasons. Firstly, the powder must be chemically stable enough during storage and shipment. The recommended T_g of powder coatings is 50°C, approximately [88]. The required T_g for the primary resin alone varies, depending on the cross-linker used with it. UV cure powder coatings can be cured at temperatures as low as 100°C [89]. Because T_g is higher, powder coatings can attain good hardness at lower cross-link density than most liquid coatings designed for similar applications. The higher T_g tends to be a favorable attribute of powder coatings, which often combine excellent hardness with excellent impact resistance [90].

The influence of the chemical structure of acrylic resin resulting in an increase in T_g can be achieved via:

- Short side chain effect is more pronounced for methacrylic esters. The methyl group located on the same α carbon atom limits the mobility of the chain. The lower mobility of the substituents increases the rigidity of the main chain, thereby increasing the hardness of the acrylic resin [91].
- The aromatic groups inside chains (e.g., styrene) generate high glass transition temperatures due to the association effect of π -electron systems in the aromatic ring structure [92].
- The effect of more polar group containing acrylic resin such as: hydroxy-functional monomers lead to higher glass transition temperatures due to high polarity effect. Other polar monomers, such as acids, amides, and nitriles, yield high glass transition temperatures also due to the additional scope for molecular association of their polar groups [93].

On the other hand, the powder during melting in oven must flow and form a desirable film. The T_g can decrease by using long chain acrylic monomers to the resin. Up to a certain point, as the length of carbon in the side chain increases, the flexibility increases. Long, aliphatic side chains on the monomers impart plasticity to the resultant polymers and generate flexible coating films. They are beneficial in respect of solubility, low solution viscosities, substrate and pigment wetting, flow and levelling, and high gloss [94].

The exceptions constitute monomers like: tert-butyl esters, cyclohexyl esters, isobornyl esters of acrylic acid or methacrylic acid, where the effect is more pronounced in

the case of acrylic esters. The side chains mentioned above contribute through their structure to the increase in glass transition temperature and influence the solubility leading to low solution viscosities [27]. For these reasons, the acrylic resin consists of a mixture of different monomers to produce the optimum balance of film properties.

THE IMPACT OF ACRYLIC MONOMER ON HARDENING MECHANISMS AND FUNCTIONAL PROPERTIES

Acrylic resins used in powder coatings classified based on the type of functional group majorly determines the choice of crosslinking agent and the curing process. The chemical reactions between the functional groups in the resin and the crosslinking agent result in a different chemical structure, leading to various properties in the finished powder coating. The choice of curing agent and the chemical reactions involved also determine the curing technology used for powder coatings. Low-temperature curing, and UV-curing technologies are examples of different methods used depending on the type of resin and crosslinking agent. Low-temperature curing is particularly useful for temperature-sensitive substrates, while UV-curing offers rapid curing and energy efficiency [95, 96]. Acrylic resin within powder coatings can contain hydroxyl, carboxyl and glycidyl functional group. The functional groups present in acrylic resins provide different curing options and lead to powder coatings with various properties, making them suitable for a wide range of applications in different industries.

Crosslinking of hydroxy-functional acrylic resins

One of the most common methods of curing acrylic resins containing a hydroxyl group is reaction with polyisocyanates. The hydroxyl groups of acrylic resins can react with isocyanates to form urethane groups. Low-molecular polyisocyanates are toxic, have high vapor pressures at ambient temperatures, and the vapors are corrosive to mucous membranes, and are thus deemed harmful. Consequently, so-called blocked polyisocyanates were developed. Regardless, such adducts must be stable at ambient temperatures. The common temperatures for urethanes formed from isocyanates and primary alcohols are well above 200°C [97]. For this reason, the special partner compounds for isocyanates with significantly lower decomposition temperatures were synthesized. The most widely chosen blocking agents are phenols, oximes, amides, imides, imidazole's, pyrazoles, 1,2,4-triazoles, hydroxamic acid esters, and active methylene compounds [98, 99]. Czachor-Jadacka *et al.* prepared blocking agent based on acetone or butanone oxime, which decrease their deblocking temperature even to 150 or 160°C depending on the pertinent blocking agent utilized [100]. The various blocking agents have different minimum temperatures for their reaction with resins

containing hydroxyl groups. These temperatures also vary with the type of polyisocyanates. For example, as aromatic isocyanates are much more reactive than aliphatic or cycloaliphatic polyisocyanates, the reaction temperatures are about 20°C lower, given the same blocking agent. The other polyisocyanates crosslinkers (PIC) blocked with ϵ -caprolactam react under above 180°C [101]. The higher the temperature, more the energy consumption, which adversely influence the production of powder coatings.

As already mentioned, acrylic resins provide excellent weather resistance, but a major constraint is the flexibility factor. The combination of acrylic resins with blocked aliphatic or cycloaliphatic isocyanates allows for optimal resistance to weather and chemical conditions and better flexibility. Nevertheless, some blocking agents cause yellowing under UV light e.g., TDI. Such powder coatings are solely used for primers and indoor coatings (e.g., wood furniture) [102].

In addition, the properties of coatings are influenced by the hydroxyl number. The hydroxyl values of acrylic resins for polyisocyanates crosslinking play a key role in the curing process of powder coatings. The greater the number of hydroxyl groups in the acrylic resin, the greater the cross-linking density. The greater the cross-linking density, the better the properties. Typically, acrylic resins for powder coatings contain low hydroxyl values of 50 to 90 mg KOH/g [103]. They are used for two-component industrial coatings and wood finishes. Acrylic resins with higher hydroxyl values of 80 to 120 mg KOH/g are used wherever higher crosslinking densities and thus better chemical resistance is required [104].

Examples of commercial acrylic resins: SYNOCURE 878 N 60 (ARKEMA), PA003 (Human Chemicals Co., Ltd.), Uralac® P 1550 (Covestro), SETALUX® 1797 SS-70 (Allnex)

Examples of commercial curing agents: VESTAGON B 1530, VESTANAT (Evonic)

Crosslinking of carboxy-functional acrylic resins

The carboxyl group-containing acrylic resin for powder coatings can be cured by using curing agent containing hydroxyl, amino or epoxy group. Nevertheless, powder coatings containing hydroxyl and carboxyl groups are most often used in the paint industry. The reaction between hydroxyl and carboxyl groups, an ester bond is formed, which is why these coatings are usually called polyester. A typical drawback of carboxyl group-containing acrylic powder coatings is its low flexibility and poor impact resistance. For this reason, modifications have been described in the literature which aim to improve these properties. Zhou described method of increasing the flexibility of coatings based on carboxyl group-containing acrylic resin. The carboxyl group attachment to acrylic resin back-bone through long flexible side chain, which not only improved the reactivity of carboxyl group but also provided enhanced flexibility

and impact resistance [105]. Okada *et al.* described acrylic/polyester hybrid powder coating. The advantages of polyester powder coatings, both -COOH and -OH functional, are good appearance and good mechanical properties such as toughness and flexibility. Nevertheless, the drawback of these coatings was the insufficient weather durability. Thanks to the development of a hybrid acrylic-polyester powder coating, the problem of resistance to weather conditions can be resolved [106].

High acid values tend to be disadvantageous because carboxyl groups do not catalyze the crosslinking reaction. Usually, the acid value of carboxyl group-containing acrylic is about 55 mg KOH/g [107]. Carboxyl functional acrylic resins are also commercially available. They can be cured with TGIC for exterior applications or with bisphenol A epoxy resins. The latter combinations are sometimes called acrylic-epoxy hybrid coating powders and are noted for their excellent hardness, stain, and chemical resistance. They find use primarily in appliance coatings [107].

Examples of commercial acrylic resins: Acryloid® AT Series (Arkema), Joncryl® (BASF)

Examples of commercial curing agent: Primid® XL-552 (Estron Chemical, Inc.)

Cross-linking of acrylic resins with epoxy functional groups

The powder coatings containing epoxy-functional group are based on glycidyl methacrylate (GMA). These chemical moieties can be crosslinked with compounds containing primary and secondary amino groups at ambient temperatures, or with compounds containing carboxyl groups at elevated temperatures. Recently, in the literature, there are numerous reports of UV-curing powder coatings. Epoxy groups derived from glycidyl methacrylate (GMA) enable the UV curing reaction through the so-called cationic photopolymerization. Cationic photopolymerization as opposed to radical-mediated processes, are characterized by the absence of inhibition by oxygen, low shrinkage, and good adhesion, and mechanical properties of the UV-cured materials. Moreover, UV-curing method also enables application on the heat sensitive materials such as: wood, plastic and MDF panels [108]. In the cationic photopolymerization, the photoinitiation step usually uses onium salts of strong acids, for example: iodonium salts, sulfonium salts, phosphonium salts, pyridinium salts [109].

Examples of commercial acrylic resins: Uvecoat 2000, 2100, 2200 series (Alnex)

Examples of commercial curing agent: triarylsulfonium hexafluorophosphate

The crosslinking reaction that occurs makes these products having high chemical and thermal resistance, as well as excellent adhesion to the substrate, especially when the resin is polymers with polar functional groups. The epoxy powder coating imparts good adhesion and

corrosion resistance [110]. Zargarnezhad *et al.* explain oxygen transport within coatings in the presence of condensed water. These studies showed that this water-induced blocking mechanism is sufficient to suppress corrosion reactions on the underlying substrate material [111]. Halim *et al.* further demonstrated that the presence of an oxide layer on stainless steel, availability of functional groups, and structure shrinkage were the factors that affect the adhesion of the carbonaceous coating [112]. Moreover, the acrylic resin based on epoxy groups has the best adhesive properties and is compatible with various additives. Mirabedini *et al.* studied different coating formulations containing various wt.% of aluminum hydroxide (alumina) particles, TiO₂ and two types of fumed silica nanoparticles were prepared via a two-stage process. Tensile strength measurements, DMTA analyses and vertical pull off adhesion test were conducted to evaluate mechanical and adhesion properties of the powder coating samples [113]. The advantages of fast curing, high hardness, excellent resistance to chemical corrosion, enable the application of powder varnish in automotive OEM clearcoats or in the furniture industry (UV curing) [114].

APPLICATION AND PERFORMANCE OF POWDER COATINGS BASED ON ACRYLIC RESIN

As mentioned, the reaction between the functional group of the acrylic monomer and the crosslinking agent allows to obtain a finished powder coating. The curing of the coating is the result of the crosslinking reaction of the polymers [115]. During the exposure of thermosetting powder coatings, the glass transition temperature of the coating film increases with increasing cure conversion. Depending on the chemical structure and the crosslink density, often a vitrification of the coating film occurs, which reduces the mobility of the unreacted double bonds significantly and prevents further conversion. Exposure at higher temperatures thus allows an enhancement of mobility, enabling further increase in conversion and further increase of the glass transition temperature, which is also reflected in improved mechanical properties, e.g., higher hardness and scratch resistance [116]. The temperature limiting the use of acrylic resins for powder coatings is 180°C [117]. Chebil *et al.* presented that the ELIUM® resin based on the polymethacrylic exhibited a degradation stage at 180°C. For this reason, the approach to develop acrylic resin for low temperature or a UV-curing powder coating systems altogether is a great solution [118, 119]. The UV-curable systems are increasingly popular as alternative way to conventional thermal curing. The high-speed process, low energy consumption due to the operation at room temperature, and environmental friendliness contribute to increasing interest in this method.

The acrylate prepolymers can be distinguished as epoxy acrylates, urethane acrylates, polyester acrylates

[120, 121]. The choice of acrylate monomers determines good coating properties after curing. Chattopadhyay *et al.* prepared epoxy acrylates/methacrylate by reacting epoxy novolac or bisphenol-A epoxy resin with acrylic acid or methacrylic acid. The epoxides were then characterized for thermal, thermo-mechanical, mechanical, and tensile properties. The UV curing process was enabled via the unsaturated double bonds of acrylic or methacrylic acid. The acrylate epoxy resins crosslinked instantly under exposure to UV radiation by free-radical photopolymerization. They were able to achieve highly crosslinked three-dimensional network [122]. The other methods of UV curing process is cationic photopolymerization. Under the influence of radiation, the photoinitiator present in the system decomposes into a cation, initiating the cationic polymerization reaction. Cationic photopolymerization involves the absence of oxygen inhibition, low shrinkage, and mechanical properties of the UV-cured materials. Further, the cationic photopolymerization curing is fast and can occur at room temperature, usually not exceeding 5 minutes [123]. Moreover, the properties of UV cured coatings are also influenced by the choice of coating components (resin, photoinitiator, additives), exposure settings such as lamp spectrum, light intensity, and exposure time. Appropriate optimization of these parameters enables the use of powder coatings [124].

Acrylic powder coatings thus offer wide range of applications. Depending on the formulation technique, they possess enhanced exterior durability, excellent chemical resistance and outstanding smoothness and gloss. Further, high scratch resistance coatings are also required of some coatings e.g., in floor coatings and automotive topcoats. The use of powder coatings is an excellent technology advancement in the automotive and architecture coatings [125]. Compared with epoxy- or polyester-based powder coatings, acrylic-based powder coatings are qualified to meet these higher UV durability and chemical resistance requirements. For these reasons, acrylic powder coatings should be explored for any application requiring exterior durability and excellent chemical resistance.

DEVELOPMENT PROSPECTS AND FUTURE TRENDS OF POWDER COATINGS BASED ON POLYACRYLATE RESINS

Based on the trends observed in the powder coatings industry, further development of acrylic resins can give more opportunity to modify them and expand their horizons in the coatings market. In the last year (in 2023) the focus was on several important topics for the powder coatings market, including:

- how to powder coat nonconductive and high temperature sensitive substrates,
- new architectural powder coatings,
- curing best practices to save money and improve performance.

Improved powder coatings formulations to revolutionize low-temperature cure capabilities [126]. Referring to the above topics, the powder coatings based on the polyacrylate resins curing by using UV-radiation or in low-temperature process, can be applied not only to metal, but also to wood, MDF boards and polymer composites [121–124]. The use of such curing techniques allows for a reduction temperature, which results in reduced energy consumption (lower costs). The selection of (met)acrylic monomers with different chemical structure allows the design of powder coatings with appropriate properties, which meet the high requirements set by customers. With the increasing emphasis on sustainability, there might be a shift towards the development of bio-based or renewable source-derived acrylic resins for powder coatings [127]. Moreover, the presence of a double bond may pose a challenge for researchers to develop self-healing or smart acrylic powder coatings [128].

CONCLUSIONS

This review highlights the significant benefits and potential of acrylic resin-based powder coatings. The investigation into the correlation between chemical structures, curing behavior, and properties of acrylic powder coatings underlines the importance of selecting the right parameters to achieve the desired application and performance characteristics. The diverse range of available acrylic monomers allows tailoring of the properties of the coatings according to specific application requirements. Compared to other resins, acrylic resin stands out for its superior outdoor performance, making it increasingly popular in the powder coatings market. Moreover, the versatility of acrylic resin enables the use of innovative technologies like low-temperature curing and UV-curing, expanding the range of compatible substrates to include heat-sensitive materials such as wood, plastic, and MDF panels. Despite its advantages, acrylic resin does have drawbacks, such as higher cost and incompatibility with certain common powder coating types. However, the exceptional outdoor properties and high performance of acrylic powder coatings often justify the additional cost.

Furthermore, the composition of acrylic resins complies with VOC regulations, making them an environmentally friendly choice. Their non-toxic nature, cost-effectiveness, ease of application, and economic benefits make acrylic resin-based powder coatings a compelling option for various applications. By understanding the relationship between acrylic resin chemistry and powder coating formulations, this review emphasizes the untapped potential of acrylic resin-based powder coatings. This comprehensive perspective provides valuable insights for researchers, coating manufacturers, and industrial stakeholders, paving the way for continued advancements in the field of powder coatings. As demands for improved coatings systems persist, acrylic resin-based

powder coatings offer a promising solution to meet both environmental and performance requirements.

Authors contribution

K.P. – research concept, methodology, investigation, validation, visualization, writing; B.P.-P. – research concept, methodology, investigation, validation, visualization, writing; R.P. – visualization, writing.

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Data availability

Data will be made available on request.

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