

Polyamidoamine Epichlorohydrin (PAAE) Wet-strength Agent: Generations, Application, Performance, and Recyclability in Paperboard and Linerboard

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Polyamidoamine epichlorohydrin (PAAE) is the preeminent permanent wet strength additive used in papermaking. Wet strength additives are used to improve paper resistance to a rupture force in wet environments. The invention of PAAE in 1957 was an innovation, as it improved paper properties by giving superior wet strength in humid or wet conditions. It was rapidly adopted by the industry. Despite PAAE's long history, the mechanism of PAAE interaction with fiber has not been fully understood. Therefore, fundamental understanding of PAAE mechanism needs to be investigated to improve its utilization in making sustainable paper products. These areas include an understanding of repulping methods and optimal PAAE dosages for better cost and performance. This paper investigates different generations of PAAE, the application method, and its impact on paper recyclability. Three generations of PAAE are currently on the market with at least two newer iterations under development. Critical application parameters that need to be understood include determination of the bonding mechanism, optimal dosage, and retention parameters. The main drawback of PAAE application is it makes paper recycling difficult. Several repulping methods are proposed for better recyclability and sustainability in the papermaking process.

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BACKGROUND

Paper products are networks of cellulosic fibers and additives held together mainly by hydrogen bonds, mechanical interlocking, electrostatic interactions, van der Waals forces, and interdiffusion of cellulose molecules. Due to the porous and hydrophilic nature of cellulose-based materials, fluids, such as water and grease, can break or collapse the network, leading to irreversible cockling. In a wet environment, the fiber structure absorbs fluids and the different types of forces and energies that hold the structure together become weak, resulting in sheet strength degradation. From a recycling perspective, the fibers separate more easily when wet. This phenomenon is considered a disadvantage when the sheet is being used by the consumer (lower strength when wet) but as an advantage or success when the sheet is being recycled or repulped in the mill (Rhim *et al.* 2007; Ray 2010; Schmied *et al.* 2013; Hirn and Schennach 2017; Cataldi *et al.* 2019; Yang and Berglund 2020; Bakker *et al.* 2022; Gralex 2023).

To improve paper's resistance to a force of rupture in wet environments, wet strength additives are used. The goal here is not to necessarily increase the tensile strength of the paper, but rather to maintain it or preserve it at a certain level (to prevent sheet failure) when the paper is wet. Wet strength agents are classified in two categories: temporary wet strength agent and permanent wet strength agent. With temporary wet strength agents, as much as two thirds of the wet strength measured after 10 seconds soaking persists for two hours; however, with permanent wet strength agents as much as 80 to 100 percent of the wet strength persists under the same conditions (Avis 1978; Espy 1994; Rita *et al.* 1999; Bajpai 2018; Vähä-Nissi *et al.* 2018; Kumar Jain *et al.* 2022; Francolini *et al.* 2023). Table 1 shows some of the commonly used synthetic wet strength agents in the papermaking process.

Table 1. Common Synthetic Wet Strengths Agents Used in Papermaking

Temporary Wet Strength		Permanent Wet Strength		
Aldehyde	Imine/Amine	Amine	Formaldehyde	Epichlorohydrin
Dialdehyde starch (DAS) ¹	Polyethylene imines (PEI) ⁴		Urea-formaldehyde (UF) ⁶	Polyamine epichlorohydrin (PAmE) ⁸
Glyoxal (GX) ²	Polyvinyl amines (PVAm) ^{5a}	Polyvinyl amines (PVAm) ^{5b}	Melamine-formaldehyde (MF) ⁷	Polyamide epichlorohydrin (PAE) ^{*9}
Glyoxylated Polyacrylamide (GPAM) ³				Polyamidoamine epichlorohydrin (PAAE) ^{*9}

* PAE and PAAE are often used interchangeably for the same chemical.

¹Espy 1995; Di Flavio 2013; Ondaral *et al.* 2018; Kurita Europe GmbH 2024

²Thorn and Au 2009; Xu 2001; Di Flavio 2013; Kurita Europe GmbH 2024

³Espy 1995; Di Flavio 2013; Kurita Europe GmbH 2024

⁴Darlington and Lanier 1995; Guerro and Lawrence 2003; Hu *et al.* 2005; Lindström *et al.* 2010; Storsberg *et al.* 2010; Kurita Europe GmbH 2024

^{5a}Kurita Europe GmbH 2024

^{5b}Pelton and Hong 2002

⁶Rita *et al.* 1999; Andreasson and Wågberg 2009; Kurita Europe GmbH 2024

⁷Rita *et al.* 1999; Andreasson and Wågberg 2009; Kurita Europe GmbH 2024

⁸Rita *et al.* 1999; Xu 2011; Kurita Europe GmbH 2024

⁹Darlington and Lanier 1995; Rita *et al.* 1999; Andreasson and Wågberg 2009; Xu 2011; Campbell *et al.* 2015; Kurita Europe GmbH 2024

Please note that Table 1 has been constructed from a liner- and paperboard perspective. The meaning of temporary and permanent wet strength is different for towel and tissue, and the placement of materials within this table would be different for those grades.

In general, aldehyde-based wet strength additives are known to have short term stability, since the aldehyde groups can undergo reactions that result in crosslinking and can result in gelation. Dialdehyde starch (DAS), glyoxal (GX), and glyoxylated polyacrylamide (GPAM) are aldehyde-based temporary wet strength agents (Espy 1995; Di Flavio 2013; Ondaral *et al.* 2018; Kurita Europe GmbH 2024). DAS is mainly used in acidic conditions (Rita *et al.* 1999). The additive is biodegradable, easy to repulp, and usually used in applications where water resistance is needed for a short period of time. GX is excellent to impart wet strength to paper without the aid of temperature or catalyst

(Xu 2001). The chemical does not easily adsorb to the cellulose, so it is sometimes applied to the paper after sheet formation by tub size or spray. The GX application can make paper brittle, which decreases folding endurance. GPAM can be used in mildly alkaline conditions, although its reaction with cellulose is more rapid at neutral or mildly acidic conditions. Papers strengthened by GPAM are highly absorptive, flexible, and easy to repulp; however, they have a remarkably high strength decay rate in wet environments. It is known that the aldehyde groups of DAS and GX react with hydroxyl groups of cellulose (and hemicellulose) to form hemiacetal bonds. GPAM is reported to provide wet strength by both self-crosslinking and by hemiacetal formation with cellulose (and hemicellulose) hydroxyls. (Young 1979; Mukherjee *et al.* 1990; Espy 1995; Veelaert 1996; Xu 2001; Di Flavio 2013; Hamzeh *et al.* 2015; Ondaral *et al.* 2018; Ziegler-Borowska *et al.* 2018; Pawlowska 2023).

Polyethylene imine (PEI) has been found to act as a temporary wet strength agent (Darlington and Lanier 1995; Guerro and Lawrence 2003; Hu *et al.* 2005; Lindström *et al.* 2010; Storsberg *et al.* 2010; Kurita Europe GmbH 2024) while polyvinyl amines (PVAm) can be used as temporary or permanent wet strength agents depending upon application and curing conditions (Pelton and Hong 2002; Kurita Europe GmbH 2024). They are respectively imine and amine-based polymers. These materials may be considered as permanent wet strength additives for towel and tissue grades. PEI can be used in both acidic and alkaline conditions and is biodegradable. The additive is expensive, yellows paper product over time, and is sensitive to light and heat. PEI-treated papers are difficult to repulp and many researchers advance contradictory bonding mechanisms to cellulose. Theories about ionic bonds, hydrogen bonds, sterically favored complexes, diffusion into fiber microcracks, amide group formation, and electrostatic interaction have been advanced, but none of them is convincing. In addition, it is known that linear PEI imparts less wet strength than the conventional branched PEI (Espy 1995). PVAm can also be used in acidic and alkaline conditions, but its application is more effective when the cellulose is oxidized. The oxidation of cellulose can give up to a six-fold increase in chemical adhesion to the fiber. PVAm is an effective additive and its bonding mechanism has been extensively investigated. According to Di Flavio (2013) PVAm adhesion to cellulose can be poor unless the cellulose is slightly oxidized, leading to two hypotheses: electrostatic bonding theory and covalent bonding theory. Although PVAm is classified as a temporary wet strength additive, Pelton and Hong (2002) have shown that the additive provides permanent wet strength. The additive adsorbs on oxidized cellulose surface forming strong covalent linkages with cellulose (Pelton 2014; Yang 2018). (Espy 1995; Xu 2001; Di Flavio *et al.* 2005; Thorn and Au 2009; Andreasson and Wågberg 2009; Di Flavio 2013; Thakur and Thakur 2016; Francolini *et al.* 2023).

Urea-formaldehyde (UF) and melamine-formaldehyde (MF) are formaldehyde-based permanent wet strength resins. They are low-cost chemicals and easy to repulp compared to other permanent wet strength agents. Both chemicals require acidic papermaking conditions for best performance. In addition, acid-resistant feed systems are required in the mill to prevent corrosion from these additives. Investigations of the bonding mechanism of UF and MF to the fiber revealed that these additives do not form covalent bonds with cellulose (Andreasson and Wågberg 2009). It is reported that both additives polycondense to protect or waterproof the existing bonds in the fiber network by reducing swelling in the bonded areas. Another way of describing this bonding mechanism is to think of the resin in a three-dimensional view where the resin encircles fibrils extending from the surface of adjacent fibers during the curing process. Once the resin is cured, these

adjacent fibers are held in place even if there are no covalent bonds between those fibrils. UF is known to reduce folding endurance; however, a paper treated with UF can retain wet strength properties for up to 8 years. MF is faster to cure compared to UF and has better folding endurance. The main drawback of UF and MF applications is the substantial amount of formaldehyde in the resins, which is a concern for safety and health. Clinical evaluation of workers handling melamine formaldehyde resin revealed pulmonary effects, decreased chest expansion, increased hilar shadows, and allergic skin reactions. Almost all papermakers have opted out the use of formaldehyde based wet strength resins for safety reasons (Young 1979; Mukherjee *et al.* 1990; Srivastava *et al.* 1992; Rita *et al.* 1999; Xu 2001; Thorn and Au 2009; Andreasson and Wågberg 2009; Roberts 2012; Zhou *et al.* 2018; Francolini *et al.* 2023).

Polyamine epichlorohydrin (PAmE) and polyamidoamine epichlorohydrin (PAAE), not to be confused with dimethylamine-epichlorohydrin that is commonly used as a source of cationic charge for balancing wet-end colloidal systems, are permanent wet strength resins that can be used under neutral and alkaline conditions (Darlington and Lanier 1995; Andreasson and Wågberg 2009; Xu 2011; Campbell *et al.* 2015). They are sometimes categorized together as polyamide-polyamine epichlorohydrin. These chemicals have good retention during the papermaking process which leads to superior wet strength performance. Compared to PAAE, PAmE is ten times less consumed in the world (Rita 1999). Its bonding mechanism is the same as PAAE bonding mechanism; however, the wet strength efficiency of PAmE resins is lower than PAAE resins. PAAE resins are easier to control due to their pH flexibility, but they are difficult to repulp. The bonding mechanism relies on the azetidinium group of the resin, which reacts with the carboxylic group of cellulose. Compared to UF and MF, the use of PAAE is credited to having no tendency towards embrittlement when the sheet ages due to the use of alkaline papermaking conditions. UF and MF require acidic curing condition which causes embrittlement of paper (Mukherjee *et al.* 1990). One of the main drawbacks of the PAmE, PAAE, or the combination polyamide-polyamine epichlorohydrin is the presence of 1,3-dichloropropanol (1,3-DCP), 3-monochloropropan-1,2-diol (3-MCPD), and organic halogen compounds (AOX), which are classified as toxic. Researchers have worked to considerably reduce these hazardous materials in the resins resulting in multiple generations of PAAE being available on the market (Mukherjee *et al.* 1990; Espy 1995; Rita *et al.* 1999; Xu 2001; Andreasson and Wågberg 2009; Diack 2015; Han and Zhao 2016; Francolini *et al.* 2023).

Synthetic wet strength resins [Table 1] are mainly used in the paper industry, which has a global market size of \$1.4 billion in 2022 and is estimated to reach a valuation of \$2.7 billion in 2031. It is important to mention that there are also natural wet strength agents such as modified starch, chitosan, modified cellulose nanofibrils, soy protein, and lignin. Although they are biodegradable, non-toxic, and environmentally friendly compared to the synthetic wet strength agents, natural wet strength agents may have performance limitations and, in some cases, are more expensive (Astute Analytica 2023; Francolini *et al.* 2023).

Synthetic and natural wet strength additives can be modified or used in combination or added to polymers to produce desirable paper properties. When using these wet strength agents, a preliminary study is recommended to determine the proper ratio or dosage needed for targeted effects in specific paper products such as facial tissues, paper towels, coffee filters, butchers' wraps, carrier boards, trays, paper plates, industrial filters, map papers, currency papers, *etc.* Although the wet strength agents are used to impart strength in wet

environments, studies have shown that these additives can also improve the dry strength of paper (Sharma and Deng 2016; Kim *et al.* 2019; Solenis Technologies 2019; Liang *et al.* 2020; Zhao *et al.* 2022; Marcello and Salam 2023; Francolini *et al.* 2023).

PAAE is the most used wet strength agent in the world. The additive accounts for more than 80% of the wet strength agents market share (Xu 2001; Diack 2015; Francolini *et al.* 2023). Understanding the different types of PAAE generations and the chemistry behind PAAE application is critical to control cost, make quality products, meet customers' expectations, and innovate or design sustainable products.

GENERATIONS

Invention

The first laboratory batch of PAAE was made on January 8th, 1957, by Gerry Keim and Ed Meginnis of Hercules Powder Company. The product was formed by polycondensation of adipic acid and diethylenetriamine (DETA), followed by alkylation with epichlorohydrin. The resulting chemical was initially called Kymene™ 557 wet strength resin, and a patent was granted and assigned to Hercules Powder Company on February 23rd, 1960 (Keim 1960a,b; Espy 1990).

PAAE is not only a cationic thermosetting chemical that adheres to the anionic fiber but also an alkaline-curing wet-strength resin. The chemical was an innovation, as it improved paper properties by giving superior wet strength in humid or wet conditions. Under the permanent wet strength additives umbrella, PAAE became a great substitute for UF and MF due to reduced machine corrosion, better absorbency, and higher wet strength of the treated sheets. Examples of paper grades that benefited from the early application of PAAE are tissue and towel products, followed by liquid packaging board, tea bag and currency. Today, although PAAE is used mainly in paper products and molded pulp products, studies have shown that it can be used to protect antibody bioactivity on paper-based bio diagnostics (Huang *et al.* 2017; Crisp and Riehle 2018; Valencia *et al.* 2020; Qin *et al.* 2022; Astute Analytica 2023).

Production

The industrial production of PAAE relies on the basic principles developed by Keim and Meginnis. The production involves three main reactions, as shown in Fig. 1. The first reaction consists of reacting diethylenetriamine to adipic acid, which leads to the formation of water-soluble poly(aminoamide). Next, the poly(aminoamide) reacts with epichlorohydrin between 20 to 40 °C to form a tertiary aminochlorohydrin. Lastly, the temperature is raised between 60 to 80 °C to promote the cyclization of the aminochlorohydrin functionality to the azetidinium functionality (3-hydroxyazetidinium group), and the resulting product is further partially crosslinked to produce PAAE resin; sulfuric acid is generally used to quench the reaction (Espy 1990; Andreasson and Wågberg 2009; Crisp and Riehle 2018; Francolini *et al.* 2023).

The final product is stable at room temperature and is usually shipped to the papermill at 12.5 to 25% solids. The main global market players of PAAE are Seiko PMC Corporation, Solenis, Ecolab, Chang Chun Group, Korfez Kimya, Buckman, and Kemira (Han and Zhao 2016; Astute Analytica 2023; Cognitive Market Research 2023).

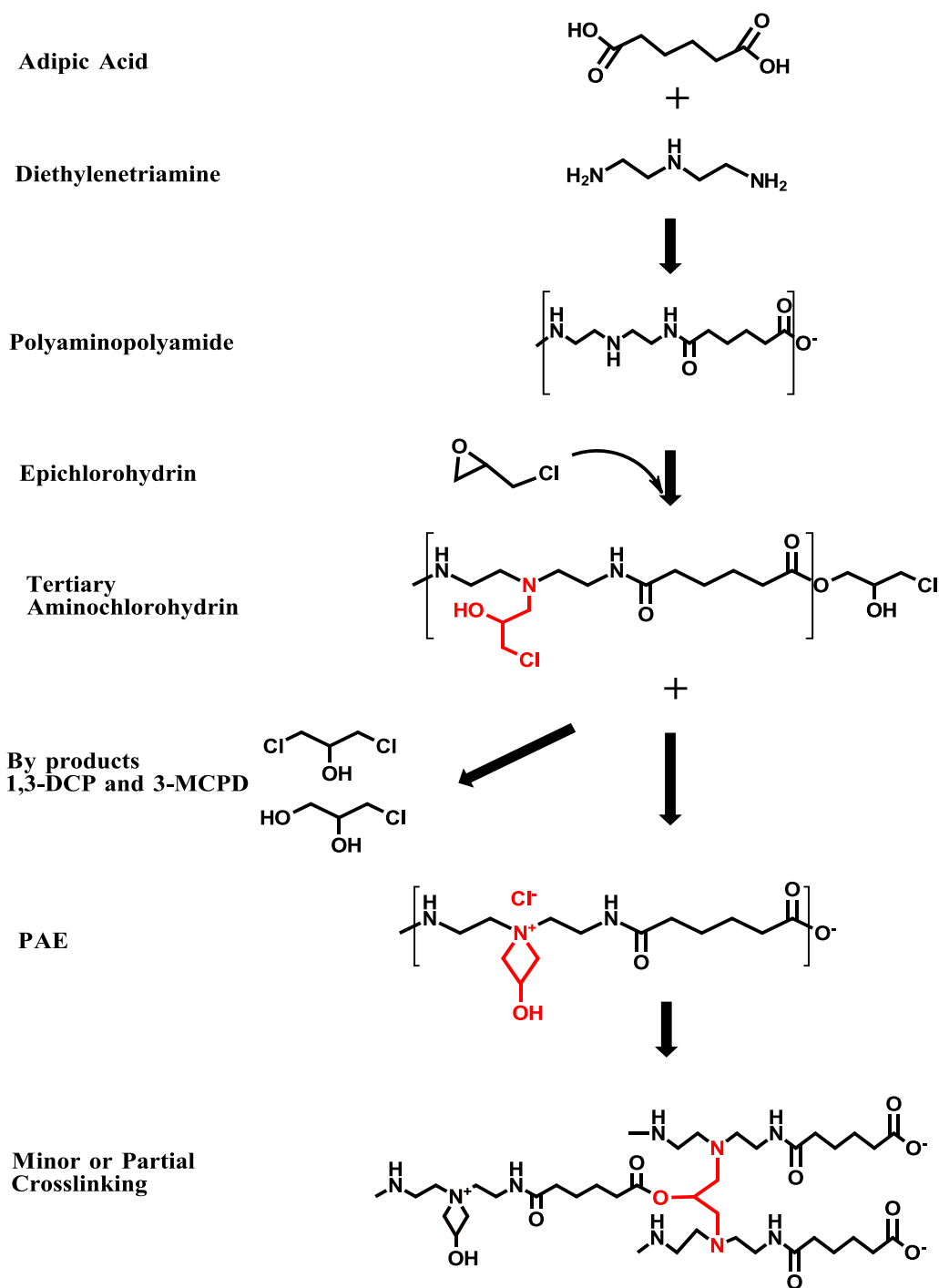


Fig. 1. General steps of PAE production

During the second and the final phase of PAE production, two main byproducts are formed: 1,3-dichloropropanol (1,3-DCP) and 3-monochloropropane-1,2-diol (3-MCPD). These byproducts are hazardous and have instigated calls for regulation limiting the amount of PAE applied to the sheet for food contact paper grades and improvement of the wet strength resin formulation to reduce or eliminate the amount of by-products being produced (Bodén *et al.* 1997; Integrated Laboratory Systems 2005; Morry *et al.* 2010; Pace 2011).

Regulations and Improvement

Despite its positive impact in the papermaking industry, the wet strength resin contains side products that are harmful to consumers. Besides adsorbable organic halogens (AOX), the main hazardous byproducts are 1,3-DCP and 3-MCPD. The 1,3-DCP and 3-MCPD are classified as carcinogens. Government agencies and non-governmental organizations (NGOs) across the world are concerned about unsafe byproducts in the resins. The German Federal Institute for Risk Assessment (Bundesinstitut für Risikobewertung or BfR), the U.S. Food and Drug Administration (FDA), the Brazil National Health Surveillance Agency (Agência Nacional de Vigilância Sanitária or ANVISA), and the National Standard of China (Guojia Biaozhun or GB) have issued government regulatory directives to curb the levels of 1,3-DCP and 3-MCPD in PAAE. In addition, globally active NGOs such as European Union Ecolabel, Blue Angel, Nordic Ecolabel, and Green Seal have called for safer wet strength resins. All these institutions, especially BfR, claim that epichlorohydrin byproducts in PAAE can enter the food chain by migration from food contact paper materials. The main BfR recommendation was to limit the extractable 1,3-DCP in water at 2 µg/L and 3-MCPD at 12 µg/L for 40 g per liter of most paper grades except for kitchen towel, for which the weight was set at 4 g per liter for the same extractable limits of 1,3-DCP and 3-MCPD (Committee of Experts on Materials Coming into Contact with Food 2009; National Health and Family Planning Commission of the People's Republic of China 2016; Crisp and Riehle 2018; Merkel 2019; The European Consumer Organisations 2021; SGS 2022).

Before 1990, the average concentrations of 1,3-DCP and 3-MCPD in the 1st generation of PAAE were respectively 12500 ppm and 2500 ppm. Post 1990s, due to governments and NGOs requirements, research and development efforts from the PAAE manufacturers led to production of the 2nd generation PAAE with the concentrations of 1,3-DCP and 3-MCPD less than 1000 ppm each. By the mid-1990s, the 3rd generation of PAAE was developed with 1,3-DCP and 3-MCPD concentrations less than 10 ppm each. It is important to note that the cost of the resin also increased as its quality improved over time. Today, to improve the cost efficiency of PAAE resin application, manufacturers are trying to produce high solid content resins (more than 25% solid) to minimize transportation, which not only reduces freight cost but also reduces carbon footprint (Crisp and Riehle 2018; Winkel 2018). Private discussions with vendors have indicated that at least 2 more generations of PAAE are currently under development. Continuous improvement and innovation may lead to harmless PAAE in the future.

APPLICATION AND PERFORMANCE

Bonding Mechanism

The PAAE chemical structure has several functional groups such as terminal carboxylic acid, amine, and azetidinium groups. The azetidinium and the amine groups promote the adsorption of PAAE onto the fiber surface. The azetidinium group which is the quaternary ammonium functionality in the PAAE molecule (Sarraf and Staib 2014) is the key element in the bonding mechanism between the PAAE resin and cellulose. PAAE reactivity arises from the cationic azetidinium group that reacts with the anionic carboxylic group on the fiber. During the drying and heating process, the azetidinium group undergoes two types of reactions which lead to the formation of an ester bond between PAAE and cellulose, or the resin can crosslink with itself using the azetidinium group, the carboxylic

group, and the amino group in its molecules. Both paths, resin-fiber and resin-resin bonding, can impart wet strength to the sheet. Figure 2 shows the covalent bonding formation between the PAAE resin and cellulose (path a) and the self-crosslinking paths (paths b and c) of the resin (Obokata *et al.* 2005; Yoon 2006; Obokata and Isogai 2007). From path a, one can deduce that it will be difficult to wet-strengthen low-yield pulp, due to the low level of carboxylic groups in the pulp.

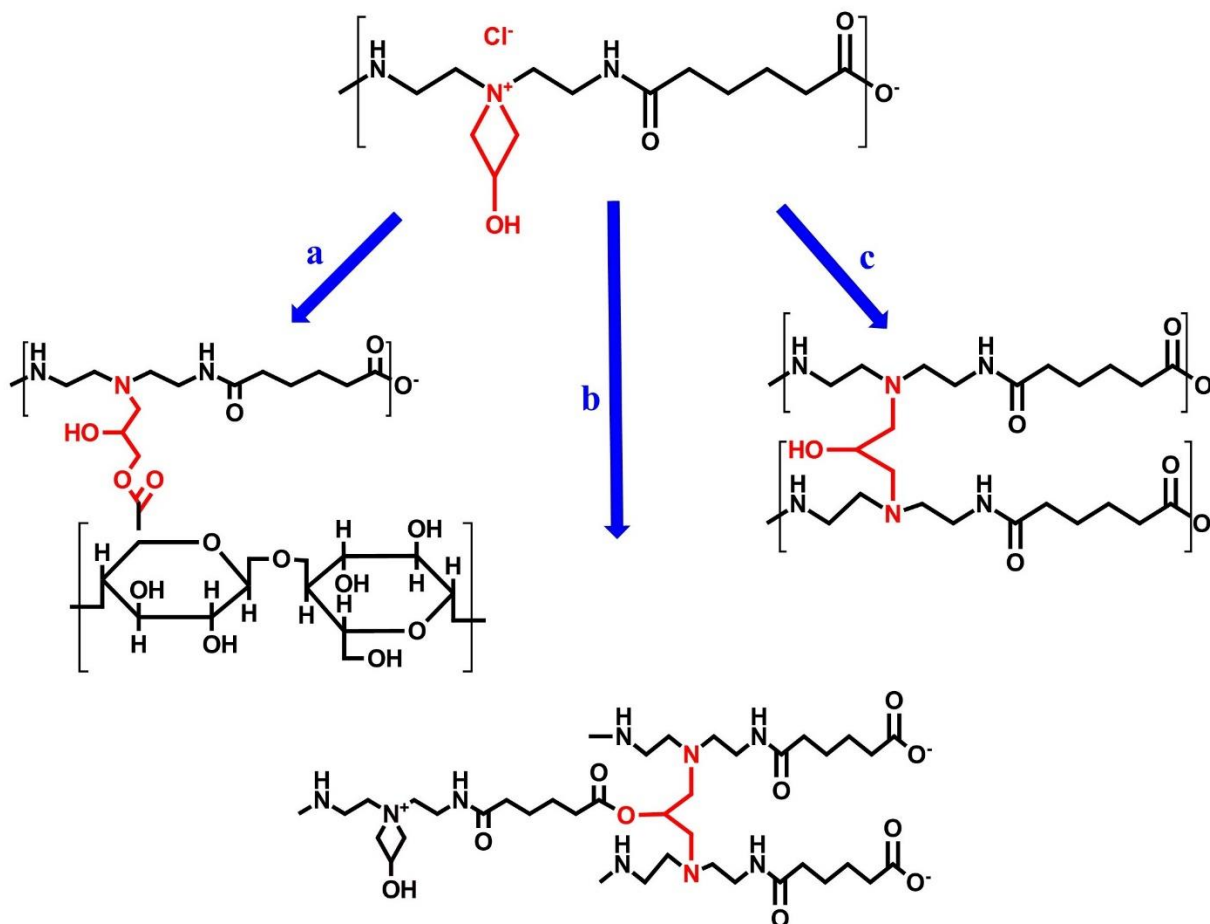


Fig. 2. Structure of PAAE (top), PAAE forming ester bond with cellulose (path a), PAAE main cross linking (path b) and PAAE secondary cross linking (path c)

While there are many factors that contribute to the development of sheet wet strength when PAAE is added to the pulp, four main factors include:

- Fiber types
- Concentration of the azetidinium group
- Adsorption time
- Drying conditions

The availability of carboxylic groups on the fiber, usually introduced during the kraft pulping process or during the bleaching process, facilitates the electrostatic attraction between the anionic carboxylated group of the fiber and the cationic azetidinium group of PAAE. A substantial proportion of the carboxylic acid content on typical fibers is due to

the presence of hemicellulose. Two phases determine the adsorption time of PAAE to the fiber: the fast dynamic adsorption phase and the slow near equilibrium adsorption phase. In the first fast dynamic phase, the majority of PAAE molecules attach to the fiber within a few minutes. During the second phase, attachment of the remaining PAAE takes a much longer time, as the fiber surface has become somewhat saturated. While PAAE resins cure at ambient temperatures (*e.g.*, TAPPI standard conditioning at 50.0% relative humidity and 23.0 °C) to provide wet strength, curing at high temperature (80 to 150 °C) leads to even higher wet strength with more covalent bonds that are difficult to break in water. It is also believed that other types of interactions might contribute to the curing of the resin, but the contribution of these interactions is less dominant (Liu 2004; Obokata *et al.* 2005; Yoon 2006; Obokata and Isogai 2007; Su *et al.* 2012; Onur *et al.* 2019; Weifang Huapu Chemical Co. 2021; Korpela *et al.* 2022).

Dosage and Performance

Vendor recommendations and mill practice is to apply wet strength resin at the wet end, preferably to the thin stock or close to the fan pump. Theory would suggest that the cationic resin needs to be well retained on the long fibers, rather than cellulosic fines. The proportion of fines tends to be higher in the thin stock. Therefore, it follows that PAAE should be added to the thick stock. The dosage of PAAE is generally 0.3 to 1.5% of the oven dry weight pulp. The PAAE dosage correlates with the strength of the sheet; however, the strength plateaus when the dosage reaches a certain limit. Physical testing such as wet tear index, dry tear index, wet tensile index, and dry tensile index are done to determine the optimum dosage. PAAE is also combined with other additives such as GPAM, bentonite, or starch for better tensile results. The addition of PAAE to the paper making process can increase the wet-to-dry strength ratio up to 35% (Andreasson and Wågberg 2009; Su *et al.* 2012; Xing *et al.* 2012; Zhang *et al.* 2017; Liang *et al.* 2020; Husić and Botonjić 2023).

There are several conditions that impact the reactivity of PAAE with the cellulose surface. These include the presence of calcium ions in the wet end, chlorine-based oxidative bleach, the ionic strength of the solution, the overdosing of PAAE and how long PAAE is stored at the mill site.

Su *et al.* (2012) demonstrated that the adsorption of the PAAE to the fiber can be enhanced or worsened by salt (NaCl, CaCl₂) concentration in the pulp. A small amount of salt allows the PAAE macromolecules to have a less extended conformation and generally promotes the interaction between PAAE molecules and the fibers (Fig. 3-B).

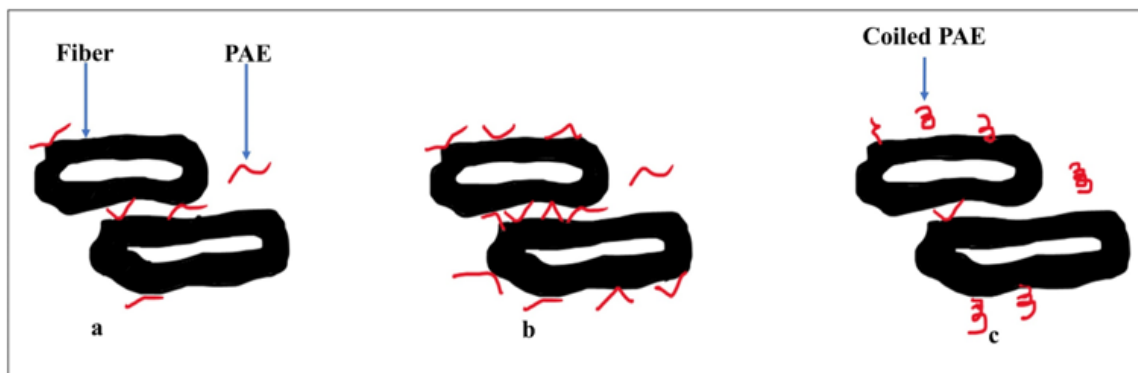


Fig. 3. Possible adsorption mechanism of PAAE to fiber at different concentrations of salt a: without salt, b: low concentration, c: high concentration. Adapted from Su *et al.* (2012)

A higher amount of salt does not promote the adsorption of the PAAE to the fiber, as too many cations in the slurry start competing with the azetidinium group (Fig. 3-C); the macromolecules become excessively coiled. In addition, calcium ions decrease the efficiency of the PAAE more than sodium ions. The excess of these ions reduces the anionicity of the fibers. Potentially, another contribution to this effect is that, in the absence of salt, the adsorption of a molecule of PAAE would inhibit adsorption of another one next to it. Salt addition reduces such charge-charge repulsion effects.

High dosage of PAAE does not necessarily translate to high wet strength. PAAE is a cationic wet strength agent, and the maximum amount of the polymer that can be adsorbed to the fibers depends on the availability of the carboxyl groups (anionic charges). When the fiber is saturated, additional PAAE may be physically entangled in the pulp but most of the extra resin will be lost to the white water. To prevent such situations, the fiber can be refined to expose more carboxylic groups for better PAAE retention. The refining solution may work, but it may introduce more fines to the pulp, thus reducing the drainage and the speed of the paper machine. There are technologies such as bipolar activator technology (carboxymethylcellulose grafting) and multilayering technology to successfully increase the amount of wet strength resin to be adsorbed to the fiber surface. The addition of carboxymethyl cellulose (CMC) enhances the amount of the carboxylic groups in the pulp, which in turn improves the retention of the wet strength resin. With the multilayering approach, opposite charged polyelectrolytes are adsorbed onto the fiber surface, which in turn favors large amount of PAE retention (Smith 1992; Hawes and Doshi 1986; Lindström *et al.* 2005; Sandberg and Andreasson 2004; Gharekhani *et al.* 2015; Fischer *et al.* 2017; Mandlez *et al.* 2020). In addition, oxidizing the fiber or oxygen delignifying the fiber will increase the availability of carboxylic groups, which will enhance the formation of PAAE-fiber bonding (Hirosawa *et al.* 2001; Dang 2007; Xu *et al.* 2022). The method for multilayering was not reported in these literature sources and hence, cannot be described here.

Behind PAAE application, it is critical to control cost, make quality products, meet customers' expectations, and innovate or design sustainable products. Vendors will often try to differentiate their PAAE products by molecular weight, charge density, reactivity, and product solids. As several of these product parameters are proprietary to the specific vendor, it is not possible to include them in this work. In some instances, one type of product will work better than another. In general, the vendors have a good understanding of the best operating conditions for each of their product offerings, and consultations with the vendor are a reasonable method for optimizing product performance.

PAAE is a self-reacting resin. Poor storage conditions with high temperature and/or excessive time results in azetidinium conversion to aminochlorohydrin and aminoglycol (reaction with chloride and water, respectively). While viscosity typically decreases with aging, self-crosslinking to gelation does periodically happen. The shelf life is up to 6 months at 5 to 35 °C. It is recommended to store the chemical in a cool dark place, away from oxides and organics peroxides (Ascent Chemicals; Toyo Roshi Kaisha 2019; Gao *et al.* 2019).

Lastly, knowing the mass balance of PAAE around the sheet is a better way to determine whether the application is efficient or not. In general, operators in the mill know the quantity of PAAE applied to the pulp but have no idea how much is retained in the paper and how much is lost to the white water. There are analytical approaches that help to determine the amount of PAAE present in the pulp or retained in the sheet or lost to the white water. Charge demand titrations can confirm that there is an excess of cationic

material in the wet end, but that is “normal” when making some wet-strength grades. Excessive addition of PAAE will often result in foam formation in the wet end.

Existing PAAE Testing Analytical Methods

Several analytical techniques such as Fourier-Transform Infrared Spectroscopy (FTIR), Nuclear Magnetic Resonance (NMR), and chromatography exist to determine the presence of PAAE. Some of these methods produce better results than others. For instance, NMR is not generally a good method to measure small amounts of material in the presence of large amount of background materials. The tests are easy to perform on pure or dry PAAE film, which is often dissolved in various solvents by the analyst prior to testing. The quantification and detection of PAAE becomes complicated once the resin is applied to the sheet. The interferences of the fiber and other paper additives make the measurement difficult. There are analytical methods that are adequate whether the PAAE is pure (raw material), applied to paper, mixed with pulp, or diluted in water. These methods developed for specific applications are discussed below:

Guise and Smith (1982) developed an analytical method to determine the molecular weight of PAAE using gel permeation chromatography (GPC). The analytical technique was used on PAAE aged approximately 3 months after the date of manufacture and performed within 1 week to avoid changes that occur due to cross-linking during storage. It is important to note that this study was not done on paper products but on pure wet strength resin. The GPC method is obviously suitable for raw material analysis.

Yano *et al.* (1991) used pyrolysis gas chromatography–mass spectrometry (GCMS) to determine the presence of PAAE in paper samples by using cyclopentanone as the resin signature peak. The technique is a direct injection method of the sheet on the pyrolyzer and may not be suitable with white water or wet pulp.

Obokata *et al.* (2005) used size exclusion chromatography attached with a multi-angle laser light scattering detector (SEC-MALS) to determine the average molar mass of PAAE. The authors also used NMR to determine 3-hydroxy-azetidinium (AZR) groups content in PAAE resins, and an elementary analyzer FLASH EA1112 to determine the nitrogen content in handsheets. These two results were used to indirectly calculate the amount of PAAE in the samples. The results from these analytical approaches revealed that the retention of PAAE to the fiber is governed by the presence of AZR.

Ozaki *et al.* (2006) were able to detect PAAE in paper using a confocal laser scanning microscope (CLSM) equipped with scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDX) and photoelectron spectroscopy (XPS). The PAAE in the samples was stained with sulforhodamine and modified with osmium tetroxide (OsO₄). The CLSM was used to locate or visualize PAAE on the surface of the pulp. The SEM-EDX was used to quantify PAAE in the sheets and the XPS was used to quantify PAAE on the surface of the pulp. It is important to note that this method works when there are no other highly cationic polymers in the sample as the anionic reagent sulforhodamine will also react with these compounds, and overquantify the amount of PAAE.

Chai and Yoon (2007) used ultraviolet-visible (UV/Vis) spectroscopy to quantify PAAE in white or bleached pulp at the wet-end. The method is not used on dry sheets, and it is not known whether sheet extraction, especially with unbleached (brown) kraft sheets, would be effective. Although UV/Vis methods are commonly used, they can be inaccurate and unreliable (Invitrogen 2018).

Siqueira (2012) carried out qualitative analysis of PAAE before and after thermal treatment in an oven using FTIR, and ¹H and ¹³C NMR. The analytical techniques could

determine the difference between rigid or flexible PAAE films after thermal exposure. In addition, FTIR and NMR were able to detect impurities such as polyamideamine (PAA) and 3-chloro-2-hydroxypropyl group in PAAE. FTIR and NMR analytical approaches were used to demonstrate that PAAE reacts with CMC when carboxylic acid groups are present in great amounts.

Ntifafa *et al.* (2023) used pyrolysis GCMS thermal desorption to quantify PAAE in paper and white water. PAAE in the samples was hydrolyzed to adipic acid, which in turn was derivatized to diethyl adipate before quantification on the GCMS. The analytical approach can be used to determine the amount of PAE retained in the sheets and the amount lost to the white water.

These analytical methods can be used to qualitatively and quantitatively study PAAE as a raw material. In addition, these testing methods can help to study and understand the retention rates of the wet strength resin in the sheet. An efficient application of PAAE not only improves the quality of the sheet but also reduces the waste of PAAE that drains with the white water during the papermaking process.

RECYCLABILITY

Proposed Recycling Approaches

The definition of paper recycling is ambiguous, as there is no clear or standard definition of the term or concept. Some examples of paper recycling definitions are “the circular process of turning old wastepaper into new paper” (Recycling.com 2023), “the process of recovering waste paper and remaking it into new products” (CTCN 2023), “the environmentally friendly process of recovering and processing scrap and waste paper to create new paper products” (Recycling Inside 2023), or “the processing of scrap paper or other such recoverable waste paper into reusable products. Such collection and recycling of recoverable wastepaper shall be done in an environmentally acceptable manner” (Law Insider 2023). The Glossary of Paper Terms (Neenah 2023) states that there is no universally acceptable definition and requirements vary by specific circumstances. Although the definitions tend to promote circular economy, sustainability, and a safe environment, they are obviously not standardized, and the procedures or methodologies to recover the fibers are not defined.

In 2022, the definition of recyclable became a contention point in the draft of the European Union packaging law. The European Commission was pushing for high target of 95% of packaging weight must be suitable to recycling for it to count as recyclable; at the same time, paper manufacturers were concerned that products will not meet the threshold due to the presence of water-soluble materials such as starch, which account for more than 5% of the paper weight in several products (European Bioplastics 2022; Taylor 2022; Commission Staff 2022; Duncombe *et al.* 2022; Committee on the Environment 2023). An objective analysis of this definition shows that yield or the amount of recovered fiber is the main factor of recyclability. In the US, the CFR (Code of Federal Regulations) broadly defines recycling as the process by which recovered materials are transformed into new products (Code of Federal Regulations 2023).

There is a need for standards that define recyclability in papermaking. A standardized definition will help scientists, researchers, environmentalists, and even lawmakers to better classify paper products across the globe. As of now, the definitions are diverse. A definition accepted in one country or one region may be invalid in another one.

In this review, recycling is defined as the process of converting back a paper-based product to pulp that can be used to make new paper products. The definition is close to that of the Pulp and Paper Dictionary, which defined recycling as the “use of recovered wastepaper and board by paper mills to produce paper and boards” (Pulp and Paper Dictionary 2023). In the mill, paper is qualified as recyclable if it is repulpable. The yield during the repulping is affected by constraints such as coarse rejects, flake content, and even macro-stickies that prevent full recovery of the fibers. Depending on the paper grade and the technique used to recycle the paper, the repulping yield can be over 80% (Paperonweb 2023; Harler 2004; Runte *et al.* 2015).

Traditionally, paper mills recycle paper products by exposing them to water and mechanical shear force in hydro-pulpers; chemical aids and/or heat are sometimes added to the slurry to break down the fiber network into individual fibers. Due to the fact that PAAE can increase or maintain the sheet tensile strength, especially when the sheet is wet, paper makers find it difficult to recycle. The pulps obtained from paper products manufactured with PAAE contain flakes of paper that are difficult to break down into fibers. Several studies were done to provide solutions to this issue (American Forest & Paper Association 2021).

Melchiorre (1997) was able to repulp beverage carrier paperboard containing PAAE using hydrogen peroxide (HP), ammonium persulfate (AP), sodium persulfate (SP), and dimethyldioxirane (DMD) at different dosages. A z-bar mixer coupled with a vibrating screen was used to liberate the fiber while not damaging the pulp. The HP application at a dosage level of 5% based on the oven dry weight of the fiber produced the lowest percent yield of 79% in 135 minutes. The dosage of DMD at 2.4% produced the highest percent yield of 92.1%. The AP and SP at 5% dosage each yielded 85% fiber in 135 minutes. The study of the handsheets made with the recovered fibers of these 4 experiments revealed that HP had the highest tensile followed by AP, DMD, and SP.

Rita *et al.* (1999) used appropriate combinations of chemical and mechanical treatment to repulp wet strength paper by depolymerizing the resin. The experiment showed that strong alkaline oxidative conditions are adequate to repulp PAAE wet-strengthened paper. The most important variables to achieve high yield are pH, temperature, consistency, time, and shear level. The optimal yield was 94% for pH greater than 11 in 60 minutes at 120 °C in a closed vessel with gentle mixing; the mixture from the vessel was transferred to a disintegrator for a shearing time of 10 minutes at 3000 rpm. The alkali used was sodium hydroxide at 7.5 g/L.

The specialty chemicals company LanXess (2023) based in Cologne markets a repulping aid called Oxone™ monopersulfate, or simply Oxone, that can be used to repulp paper products such as tissue, milk carton, carrier board, and poly-coated board that contained high wet strength resin PAAE. The supplier claims that the chemical is chlorine-free, offers fast efficient removal of PAAE, and helps to obtain both high pulp quality and yield. In 2021, The Green Seal (2021), which is a global nonprofit organization for a healthier and a greener world, assigned to LanXess the raw material verification seal for Sanitary Paper Products, Paper Products Used for Food Preparation, and Food Service Packaging.

Yang and Luetgen (2020) later studied the repulping process of commercial multifold paper towel containing PAAE wet strength agent by using the Oxone of LanXess. The results indicated that fiber recovery with the use of Oxone was as high as 88% at 2.4% dosage at 60 °C in 35 minutes. A bench-top pulper at 5% pulp consistency with a pH adjusted with sodium hydroxide to 10.5 was used during the first phase of the experiment.

In the second phase, the slurry from the pulper was disintegrated in a British disintegrator for 10 min at 2000 rpm. The final slurry from the disintegrator was screened on a Somerville screen for 40 minutes. According to the authors, Oxone not only helps to achieve high yield fiber recovery but also a high physical tensile strength, burst strength, and tear resistance.

Chatrath and Durge (2011) described repulping of wastepaper having up to 35% wet strength by using a combination of sodium hydroxide (NaOH) and polyethylene glycol (PEG). The experiment was carried out in a sealed autoclave digester at 8% consistency for 30 to 75 minutes. The pressure and the temperature were respectively 1.3 kg/cm² and 105 °C. The yield of the experiment was 95%. According to the author, using sodium hydroxide and polyethylene glycol produced less fines and gave a high yield of fibers, which favored higher freeness. A combination of 1% NaOH+1% PEG led to a freeness of 52 °SR and a combination of 1% NaOH+2% PEG led to a freeness of 55 °SR. The authors stated that similar results were obtained for an open vessel experiment.

Chen *et al.* (2012) were able to repulp high wet-strength waste banknote paper using a dual acidic and alkali approach. The soaking time, temperature, and the pulp consistency were the critical parameters to achieve higher yield. The optimal fiber recovery was attained when the sample was soaked at 10% consistency for 30 minutes in 5% sulfuric acid, washed to neutral pH with deionized water, and soaked back for 30 minutes in 5% caustic soda; in both cases, the soaking temperature was 80 °C. Next, the obtained sample was centrifuged to about 25% consistency and refined in a PFI mill at 5% consistency with 0.2 mm clearance for 4900 revolutions. The final pulp consistency was adjusted to 0.5%, and a slotted screen with a width of 0.30 mm was used to estimate the pulp yield. The optimal yield was 88%.

The work of Su *et al.* (2012) showed that the success rate of the repulpability of wet strengthened paper correlated inversely with the amount of wet strength agent initially charged to the paper. According to the authors, the recyclability index, which is the percentage of fiber satisfactorily repulped and recovered, sharply dropped from 95% to 58% when the dosage was increased from 1 to 2 mg PAAE/g of fiber. In their study, the Messmer laboratory disintegrator Model MKIIC (Messmer Instruments Ltd, Gravesend, United Kingdom) was used to disintegrate a shredded 1.2 g handsheet in 2 liters of deionized water for 3000 revolutions in one minute. The experiment was carried out at room temperature. The resulting slurry was screened in a Brecht & Holl BH-6/12 fiber classifier with 0.2 mm slot screen for 20 minutes at a flow rate of 2.5 liters per minute. The study results showed that paper recyclability linearly decreased as the concentration of the PAAE increased.

The study of Yang *et al.* (2019) was focused on how to turn off the strength that the PAAE resin provides to the fibers to make the paper more recyclable and compostable. The researchers used a PAAE loaded with reductant-degradable poly (N-isopropyl-methacrylamide-co-acrylic acid) microgels as the wet strength additive. The PAAE-microgel additive was prepared by mixing 80 mg of PAAE and 60 mL of 1 mM sodium chloride solution at pH 7 for 1 hour; 10 mL of the resulting solution was added to 50 mg of the microgel, and the mixture was agitated overnight. Unbound PAAE was later isolated to recover the newly prepared wet strength additive, which is a PAAE-loaded microgel.

The PAAE-loaded microgel additive was used to treat wet, oxidized, and regenerated cellulose membranes for 24 hours at 23 °C at 50% relative humidity. The obtained cellulose membranes were treated at pH 9 with 10 mM dithiothreitol (DTT) reducing agent. Wet adhesion tests were performed to study the extent of deactivation of

the wet strength additive. The results showed about 30% decrease in wet adhesion at room temperature and up to 46% decrease in wet adhesion at 85 °C. According to the authors, the reducing agent DDT switched off the PAAE-loaded microgel. The authors believed that this innovative approach of deactivating the PAAE can help improve the recycling and composting of potential PAAE strengthening paper products. It is important to state that this work was proof of concept and was not based upon paper testing; the results are promising but not yet fully conclusive. In addition, the authors also stated that microgels are not commercial, nor green, and are unlikely to ever be economically viable for commodity paper products.

Contech has a technology to recycle or remove wet strength resin that clogs felts during the paper making process. If all the resin is not retained to the fiber, then part of it is pulled into the felt pores and eventually induces clogging. To prevent the occlusion of the pores, Contech developed products that can be used for batch and continuous felt washing processes. Although the ingredients or the active chemicals of their products are not known, probably due to trade secret, the company claimed to offer a portfolio of various products and equipment that significantly improved the removal of PAAE deposits (Veríssimo 1987).

The benefit from PAAE application is also the potential source of its main drawback. The wet strength provided by PAAE to the paper is useful; however, the same created strength becomes a significant challenge during the repulping process. The proposed repulping approaches are intended to reduce or eliminate the variability in the recycled pulp quality. Repulping aids, temperature, pH, and time can be used in different ways to achieve higher fiber recovery. In addition, the yield can be further improved by adding a deflaker to break up patches of wet strength bundled fibers during the repulping process (GreenBlue 2011).

Future of the Recyclability of PAAE Wet-strengthened Papers

It is important to note that most of the published repulping approaches of wet strengthening papers have been performed at the laboratory level. However, wet strengthening papers have been repulped at an industrial scale by several paper companies that do not necessarily publish their approaches in scientific journals. Several mills repulp wet strength agents containing materials by applying high pH, high temperature, and high shear with various levels of success. Some mills also add oxidative chemicals such as hydrogen peroxide to assist in increasing repulper fiber yield (Swales 2014). In addition, some of the methods are highly corrosive, may accelerate corrosion, or necessitate high energy to heat the water during the repulping process. Most of these studies have focused on pulp yield and quality. Less is known about the produced white-water quality, which can be a source for micro and macro stickies, high biological oxygen demand (BOD), or other sources of byproducts that may negatively affect the papermaking process. A pH swing can also be observed after the repulping process; for example, the pH dropped from 10 to 8.8 when Oxone was used to repulp PAAE based towel paper (Chatrath and Durge 2011; Yang and Luetgen 2020).

More studies need to be done to determine the acceptable PAAE-fiber ratio suitable to recycle. Studies have shown that paper charged with low amounts of PAAE are easier to repulp (Su *et al.* 2012). In addition, a combination of PAAE and other recyclable products may preserve the wet strength performance of the paper while allowing for recyclable. For example, the use of carboxymethyl cellulose (CMC) and a reduced amount of PAAE can impart high wet strength to the sheet (Christenson 2023). It is also known

that the substitution of cellulose nanofibres (CNF) made of bleached radiata pine softwood kraft pulp can enable the PAAE dosage to be reduced by over 95% while maintaining wet strength properties. Lower resin dosage not only reduces the manufacturing cost of the paper, but also reduces the contents of both volatile organic compounds (VOC) and chlorinated by-products. Lastly, the difficulties encountered in the recycling of high wet strength paper products is a call to innovate new PAAE resin that will be safe and totally recyclable (Ringold and Furman 2019; Onur *et al.* 2019; Christenson 2023).

The comparison of repulpability or recyclability rate of different wet strength resins could help papermakers or consumers to understand how the paper product could be repulped or recycled. Although, as of now, there is no available detailed or compared experiments about the repulpability degree of different wet strengthened papers, several authors mentioned which wet strength resins are easier or difficult to repulp. Pawlowska (2023) from Kemira Chemicals explained that GPAM can be easily repulped without repulping aid or excessive temperature. The same author found PAAE difficult to repulp when using water only. Kim *et al.* (2007) found it problematic to repulp papers containing PAAE, UF, and MF wet strength agents. Mukherjee (1990) found that UF and MF treated papers can be easily repulped under acidic conditions, but PAAE treated papers are not readily repulpable under acidic or alkaline conditions. According to the author, DS offers extremely easy broke recovery. Referring to Xu (2001), polymeric amine-epichlorohydrin resins such as PAAE and PAmE cannot be easily repulped. According to Dulany *et al.* (1996), a paper treated with a composition of resin made of polyamine, a polycarboxylic acid or ester, a dialdehyde and epichlorohydrin is more easily repulpable than paper treated with conventional wet strength agents. According to The Alliance for Beverage Cans and the Environment (2022), papers with temporary wet strength agents are easily recycled. The authors mainly rely on the terms “easily” or “difficult” to describe the repulpability properties of wet strengthened papers. There is obviously a need to standardize the repulpability properties of wet strength agents with degrees, scales, or more scientific terms. This can be achieved by comparative studies of several wet strengthened papers with different types of wet strength agents and different dosages to the pulp.

CONCLUSIONS

Polyamidoamine epichlorohydrin (PAAE) is a permanent wet strength additive that imparts superior wet strength to paper products. The chemical has been improved during the last decades and remains the most consumed permanent wet strength resin in the papermaking industry.

- An improved version of PAAE, 3rd generation, has extremely low organic chlorines carcinogenic compounds.
- Understanding PAAE’s bonding mechanism to the fibers can help control costs and efficiency in the papermaking process.
- PAAE is a cationic polymer that retention depends greatly on the availability of the carboxylic groups on the fiber. Refining and adsorption technologies can be used to improve PAAE retention and performance.
- The recyclability of PAAE paper products is the main drawback of its application. There are repulpability approaches to recycle PAAE wet-strengthened paper

products, although most of these approaches have only been demonstrated at laboratory scale.

- The recyclability of paper products prepared with PAAE depends strongly on the quality of the yielded fiber.
- More study needs to be done to understand the quality of white water or wastewater produced during the repulping process.
- More studies are needed to determine the acceptable PAAE-fiber ratio suitable to recycle using conventional hydropulpers or less repulping chemicals.

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