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Review

Novel Approaches to Enhancing Sustainable Adhesive System Solutions in Contemporary Book Binding: An Overview

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Abstract: This paper contributes to eco-efficient and sustainable book binding production. Higher book binding manufacturing efficiency—with less waste and reduce energy consumption—has been achieved with higher inputs of natural biodegradable sources into graphic arts materials through the eco-labeled paper grades and the use of eco-advanced adhesive system solutions. Nowadays, scientific sources on non-toxic polymers and resins, combined with current scientific knowledge and production development, are closely related to sustainability. Hence, advanced and improved adhesive system solution technologies should fulfill the needs of suppliers and customers who are involved in the International Framework for ISO/TC130 workflows. These strategic partnerships provide possibilities in the context of "closed-loop recycling models", which spark and advance the discussions of stakeholders. It is very important that the novel engineered biodegradable adhesive system solutions provide productivity-increasing and cost-effective solution performances by saving money and improving the performed binding activities. Without doubt, the task of the scientific community is to continue to provide responsive and comprehensive approaches to fulfilling stakeholders' specific needs through standardized quality assurance, with the emphasis on book-binders.

Keywords: bookbinding; adhesive binding styles; renewable adhesive system solutions; sustainable adhesive binding style solutions

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1. Introduction

The book value chain is essential since it enables access to literature and culture by improving reading habits in society. Regardless, however, digital technologies such as e-books are increasingly finding their place in society today, while print book editions played their part in the society of the past. A recent report given by the Organization for Economic Cooperation and Development (OECD) claims that students spend more time online (for school activities and entertainment), while printed books are essentially enjoyed offline [1]. However, the OECD study also reveals that young people enjoy reading more when they read in print, not on multimedia gadgets. INTERGRAF [2] and other European and national authorities recognize books as essential cultural goods and educational resources. Although the internet and television are popular media, print media still has a strong foothold amongst readers because it gives options for people to reach a varied bracket of products which have a loyal readership. Holding a physical book gives us a sense of touch, a kind of experience that is impossible to reach with e-book. Opening bookshops in local communities enables the development of critical thinking in society. Thus, bookselling, publishing, printing, and bindery sectors stand united in emphasizing the essential value of books [3]. In 2019, for the European Union, the book revenue was estimated as approx. EUR 22.4 billion with a total market value of EUR 36–38 billion [4]. The contemporary bindery sector produced about 605 thousand new book titles [4], which was a slight increase in comparison to 2018.

One of the most important steps in the production of books is the bookbinding process—the process of book formation. In general, it can be divided into edition (publishing)

binding and library binding. Edition binding definition refers to the uniform style of mass-production methods in contrast to hand bookbinding or library binding. Library binding means the reparation and restoration of works in the bindery sector. Edition binding also includes "short-run" book production, which supports self-publishing and handling binding production independently. As previously mentioned, edition binding refers to the books' binding quantity, while library binding implies typical book repairing and conservation processes.

Digital publishing through print on-demand services includes the surge in self-publishing titles in different book formats which results in the reduction of bookbinding production. In the INTEGRAF Economic Reports given for the period 2000–2023, it was reported that edition binding mass production (catalogues, books, advertising, commercial, magazines, etc.,) is less prominent in lithographic printing presses (heat set/cold set web offset, sheetfed offset) [5]. Moreover, the INTEGRAF verified a significant increase in digital laser printing and digital press processes (direct imaging press, variable data printing technology, web to print) in edition binding services on demand [5]. On-demand binding experiences are rapidly growing; for example, through using Xerox digital press technologies which produce high speed copies and can provide reprints and new short-run book editions at a reasonable price. Therefore, the bindery sector can offer fast, reasonably priced, durable, and attractive book products including different binding styles.

With regard to the global market and development of new printing machines and graphic arts materials, the graphic industry today strives for rapid development with minimal costs and ecological sustainability in view of increasing environmental awareness and the development of environmental legislation. Taking this into account, as well as the lack of scientific papers with bookbinding as the subject, the aim of this paper is to summarize the recent knowledge and to point out some potential problems occurring in bookbinding sector. In addition, the aim of this paper is to highlight the importance of using graphic materials that have an impact on the sustainability of the technological process of bookbinding. According to Towers, "new green book designing" requires the tailoring of work procedures and tools which improve the environmental efficiency [6]. The appropriate development concepts (eco-bookbinding strategies) rely on consumable materials (paper and adhesive) such as recycled paper and modified eco-friendly adhesives. Moreover, it can be concluded that environmentally friendly bookbinding includes the use of renewable feedstocks in the production of paper and adhesives, as well as other means of sustainable production such as green energy. Sustainable eco-book designing has a technological and a material component, which help reduce the impact on the environment through the reduction of the carbon footprint and hazardous waste. In the early stage of book design, the producers should also attend to increased safety in the working environment. This means that they will be knowledgeable about the materials used and their handling. This can be achieved by accounting for the use of nontoxic materials and their optimized consumption when designing the final product. In the end, producers should take care to optimize production and think in advance about product quality and durability, as well as the product's end of life solution (biodegradability or recyclability). Consequently, optimal efficiency in multiplied bindery jobs should be obtained through global stakeholder coalitions supporting local communities to reduce environment impact [7,8]. Therefore, sustainable development in the bindery sector should bring in new eco-strategies which rely on economic activities while simultaneously corresponding to the agendas of global stakeholders. Moreover, practical sustainability in the bindery sector (edition binding and/or library binding) must be up to date with regard to the sustainable aspects of production, including cleaner binding production and the provision of clean, safe working environments in the bindery sector. Sustainable book production is attained only through engineered designing, i.e., by a checklist of targets which deals with the total environmental impact of manufactured books. The targeted functions such as safety, usability, durability, reliability, and cost are listed in Designing for Environment (DfE) checklists [9]. Items such as consumption of graphic arts raw-materials, toxicity, price avail-

ability, process-related processing capability, and susceptibility to material recycling ensure sustainable development through "eco-labeled criteria" and the production of prototypes. Such concepts established in the bindery sector (edition binding and/or library binding) ensure the durability of book products and its components, as well as the restoration and upgrading of book products, easy cleaning methods, and reusability.

2. Library Binding vs. Publishing Binding

Published books for mass audiences are produced in high volume and delivered at the lowest possible cost, including restricted budgets and limited choices [10,11]. These are contemporary methods of edition binding (paperbacks, hardcovers) which are systematized by prof. Warner Rebsamen, PhD from Rochester Institute of Technology in New York [12–14], and certificated by the Library Binder Institute [15], which applied rigorous library binding Standards (LBIZ39.78-2000).

The Library Binding Standard documents monitor binders' knowledge, equipment, and experience in the process of book binding. Library bound books must incorporate high-quality graphic arts materials and rigorous manufacturing processes because of important levels of circulating books in libraries. These longer shelf-life books must also reduce expenditures for the replacement of damaged graphic arts materials through their library rebinding and failed published (edition) bindings. For that reason, the library community must work together with binders actively, which directly contributes to the development, maintenance, and restoration of books, and consequently to the development of new sustainable technical standards, quality advanced graphic arts materials usage, expertise support, and economical products usage. Thus, suppliers and technologically advanced binderies together must ensure the best suited services for customer needs. They need to understand the different binding standards, including the ones recently mentioned, which enable book products manufacturing.

Books come in different formats and binding styles and the procedures of books binding affect their price directly. The book's attraction, functionality, and durability mostly depend on the book block and cover appearance. Thus, paperback and hardcover binding types directly influence the book's durability and its manufacturing budget, while the book block binding form defines its durability against mechanical damage through insurance of easy page opening and scrolling. Hardback binding procedures imply the thread sewn book block and a stiff case made in a separate way, where the endpapers link the book block and the stiff case covered with adhesive. Hardback edition binding, including case making procedures, is always related to the book block, which is made up of signatures (folded printed sheets) as a binding unit. Undoubtedly, hardback binding types are more expensive and durable than paperback ones. The most represented paperbacks in book manufacturing are a perfect example of binding style. In this form, the loose leaves, as binding units, are tightly glued together into a book block, and the spine of the book block is binded with a soft cover. This binding style is a logical choice for many publishers due to the smaller number of binding operations, which significantly reduces manufacturing cost. Thus, paperbacks are more convenient to be carried around due to their lighter weight, but are also prone to damage, especially on the spine of a book block, which causes pages to fall out. Additionally, paperbacks are not able to be laid flat on the table like hardcover ones. In digital printing, "loose leaves" as binding units in perfect edition binding provide the opportunity for self-publishing, which is much cheaper than the hardcover alternative. Producing a book block is also much easier in perfect binding editions than in hardcovers. In addition, the perfect binding cover is made from heavy-weight cardstock paper that is laminated in order to protect the cover. Different usage of adhesives and their characteristics in perfect book binding manufacturing support performance benchmarks and provide a novel platform for additional improvement, especially with cold adhesives. Moreover, perfect binding style can only be fitted to hardcovers for presentation purposes. Nowadays, creative book-binders, experts, and suppliers are together coming up with binding method

type novelties, thanks to advanced "eco-labeled" graphic arts materials that offer unique product solutions for different book purposes and their attractive appearances.

3. The Viewpoints in the Contemporary Binding Processes with Different Adhesive Solutions

Generally, librarians' work is engaged with the physical preservation of printed materials and include only hand-binding processes such as archival restoration, job binding, design, custom binding, and making of protection boxes. The National Information Standards Organization and Library Binding Institute described in detail the technical specification for paperback and hardback manufacturing, exclusively for library use through the useful life of library books [15]. Binding production techniques and automated procedures are presented in detail, including maximized binder capability, selecting a treatment method, standardized industry practices, and material specifications. Technical specifications include methods for the first-time hardcover binding and rebinding; suitable binding methods for different binding units (loose leaf and signature); various binding methods of leaves and endpapers attachment (gluing and sewing); different methods of spine book block treatments (backing enhances, rounding, lining); and specified methods of hardcover case making (covering materials, cover boards, adhesive).

The PVAc adhesive (a polyvinyl acetate polymer water-based emulsion) is an adhesive characterized by good long-term aging characteristics for binding styles (double fan, gluing the spine, lining the spine, making the case, casing in) and it is widely applied in edition binding including on demand, short, or middle run book manufacturing [15]. However, in large run book production (paperbacks and hardbacks), PVAc adhesive does not fit well with automated technologies. The physical characteristics of the adhesive significantly prolong the opening time of bringing two substrates (adhesive and paper) together in automated workflow processes. In contrast, synthetic thermoplastic (EVA hot melt) and reactive thermoplastic (polyurethan hot melt) adhesives could be excellent choices because they ensure a higher speed of automated operations.

On the contrary, publishing (edition) binding style ensures mass bookbinding production and handmade bookbinding in particular. It further gives standardized procedures high-volume quality production of the same or similar end-products. Manufacturing concepts are identified with the increase of advanced technologies, graphic arts materials, human resources engineering, facilities, and sustainable job planning in the bindery sector in terms of project management. The concept of sustainable workflow and development must be involved and improved in the bindery management system throughout "eco-labeled book binding" procedures which reduce the impact on natural resources such as soil, water and air. Nowadays, waste management procedures, as one of the major problems of modern society, require more focus on the education activities of bindery laborers [16]. This approach could establish an adequate waste management system in which eco-qualified workers participate through advanced binding style concepts. The International Organization for Standardization created Framework for ISO/TC 130 Standards within print and publishing was brought together under the title 'Graphic Technology [17]. The Framework document is divided into two major categories which include the technical contents of each standard. The first part of framework relates to multiple graphic arts processes including all participants, while the second one relates to single graphic arts processes including job initiation, data preparation and exchange, pre-press, printing, post-press (bindery, packaging, converting), and final product packaging and logistics. The framework deals with technical issues: answering who the target users (stakeholders) are; managing interrelationships among different standards; and answering who is responsible for its updating or development. Thus, the post-press operators in the bindery sector take on multiple responsibilities according to the standardized guidelines of single binding production standard ISO 16763 [18] and the guidelines of multiplied graphic arts processes related to category standards (terminology, media and materials, measurement and testing, quality control, conformity assessment, ergonomics and safety, and environ-

ment and ecology) [17]. This workflow systematization ensures reliable results in the bindery sector. The Framework document joins process control stakeholders and suppliers, in which standardized guidelines are respected through ensuring well-defined binding manufacturing parameters (mass production, middle/short runs production, handmade production), which are integrated in logistic and environmental practices. In edition book binding (paperbacks, hardbacks), various binding forms (sewn or unsewn) of book block have been performed. According to the Framework for TC-130 Standards, advanced adhesive technologies are welcome in bindery manufacturing, especially for the unsewn perfect binding style, in which one-shot thermoplastic adhesives are used for the widest possible range of paper stocks [17]. Contrary to what was previously mentioned, the thread-sewn Otabind (Finland) binding style and the Swiss style need special equipment, in which one or two shots of the PVAc adhesive can be applied on a book block spine. Unfortunately, one-shot thermoplastic adhesive recyclability (EVA and PUR) asks for additional sustainable efforts to replace the synthetic resin with advanced natural system solutions. The PVAc adhesive is a water dispersion containing solid particles and is mostly tailored-made for different gluing solutions due to its perfect flexible performances. In edition book binding, emulsion adhesive is used in hardback book binding operations such as gluing of line strip on endpapers, gluing a strip on back of sewn book block from book's head to tail, forming the case by gluing the boards to the cover material, the insertion of the book block into the case, spine book block gluing, and side book block gluing. The PVAc adhesive is widely used in unsewn binding styles in handmade or short-run edition manufacturing because it allows a little time for work corrections [19]. Paste (starches) can be added to PVAc adhesive, although the paste can slow down the drying process; thus, the adhesive mixture performs faster-draying elastic properties [20,21]. Finally, animal adhesive, which is applied only in the case of making workflow operations, can be used in semi or line hardback manufacturing [22].

3.1. Comprehensive Guiding Principles of Adhesive Application in Bookbinding

Generally, the appropriate use of adhesives enhances the appearance of a bound book. Gluing procedures are interrelated with the following ones, and gluing deviations in the current step leads to quality deviation in the end-product. For that reason, edition binding manufacturing must be guided by standardized procedures and the prescribed tolerances of the ISO 16763:2016 standard [18]. Moreover, stakeholders (binding manufacturers and adhesive suppliers) tend to enhance overall manufacturing and assessment of bound books by enhancing production efficiency and accuracy. Above all, manufacturers need to reduce the occurrence of remakes resulting from wrong working procedures. Moreover, they need to promote quality requirements of adhesive applications in the understandable manner, particularly through additional standardized procedures described in the ISO 9706:1994 standard, [23], which ensures long shelf-life bound books. The standardized library binding procedures LBIZ39.78-2000 [15] are connected directly with operators in the edition binding sector. Both mentioned international standards encourage advanced research and possibilities to maintain other standardized procedures for pulp, paper, and board (ISO 186; ISO 187, ISO 302, ISO 536, ISO 1974, ISO 4046, ISO 5127-1, ISO 6588, ISO 10716) [24–32], ensuring chemical and physical bound-book permanence during handling. For decades, binders have coped with linking paper fibers with adhesives, especially with various fine coated paper grades, which have huge application in commercial high-speed edition binding. Modern binders need to be capable of offering a very economical style of adhesive binding service, in which the adhesive binding technologies guarantee the optimum linkage between the adhesive and the paper substrate. Nowadays, bound magazines are the top end-product in commercial bindery. Magazines consist of more than 70% of the recovered (recycled) paper because a magazine's life span is the shortest compared to other bound-book editions (catalogues, pocketbooks, handbooks, monographs, etc.). These long-span end products comprise different office paper grades; they use virgin and recycled fiber resources and non-fibrous components, such as fillers and additives, all of which guarantee

paper printing performances [33]. For multicolor printing of the office bulky paper surface, which contains a small amount of lignin in its structure, the paper needs to be coated or varnished due to specific surface properties. However, when using recycled paper in in multicolor printing, the coating procedure is obligatory due to high impurities content in paper sheets. As previously mentioned, paper fiber's linkage with adhesive depends on the paper binder's possibilities. The formation of an adhesive joint is brought by the interfacial contact between the adhesive and the substrate. Once the interfacial contact is established, some kind of intrinsic adhesion needs to take place that will hold the adhesive and substrate in place throughout their life span, with the exception being in cases where only mechanical locking occurs. There are several adhesion theories: physical adhesion, chemical bonding, diffusion, electrostatic, mechanical interlocking, and weak boundary layer. In all cases, adhesive joints involve molecules in contact, i.e., physical contact; for example van der Waals forces over the interface. Chemical bonding includes the formation of covalent or ionic bonds over the surface. Mechanical interlocking occurs between the adhesives and irregular surfaces of porous materials, when the adhesive is placed between the surface irregularities prior to hardening. Taking that in mind, paper surface is a good adhesion substrate due to its surface free energy and porous structure, providing high contact area. The strength of the adhesion joints depends on different properties of paper and adhesive. With a proper binding style, almost any paper grade can be successfully bound into a useful and durable end-product [34]. Three paper characteristics contribute to the spine structure of a book block:

- *Drapability*: this paper ability expresses the natural falling and draping of a paper sheet into the gutter margin of an opened book (paper stiffness declines perpendicularly to fiber grain direction and paper sheet bends more easily);
- *Bindability*: this paper ability expresses the successful binding with adhesive solutions and the paper cohesive strength which appears under stress conditions;
- *Cohesiveness*: this paper ability expresses a possessed paper energy on bond lines.

Bindery adhesives are made from different substances to achieve the effects with different substrates (paper, mull, fabrics, plastics, wood, leather, etc.). In general, adhesives are composed of polymers and additives (fillers, plasticizer, stabilizers, etc.) which define their application and basic properties (Figure 1).

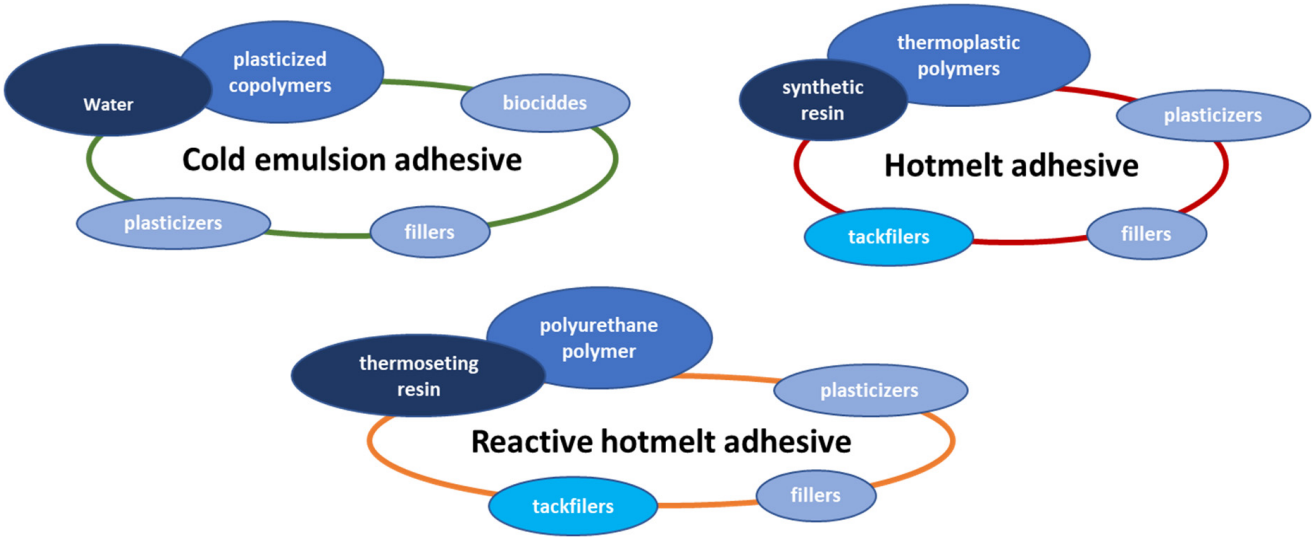

Figure 1. Basic composition of the adhesives mostly used in bookbinding.

In different book binding styles, the adhesive is a unique layer that is the observed in structure of a book block spine. Different adhesives have different effects; they typically tend to form film layers rather than the actual bond with the substrates. Quality of bindery

adhesives depend on their flexibility, their elasticity, and the thickness of a formed film layer. Flexible adhesive bends easily without breaking and returns to its normal shape after being stretched or compressed, which directly confirms its elastic properties (Figure 2).

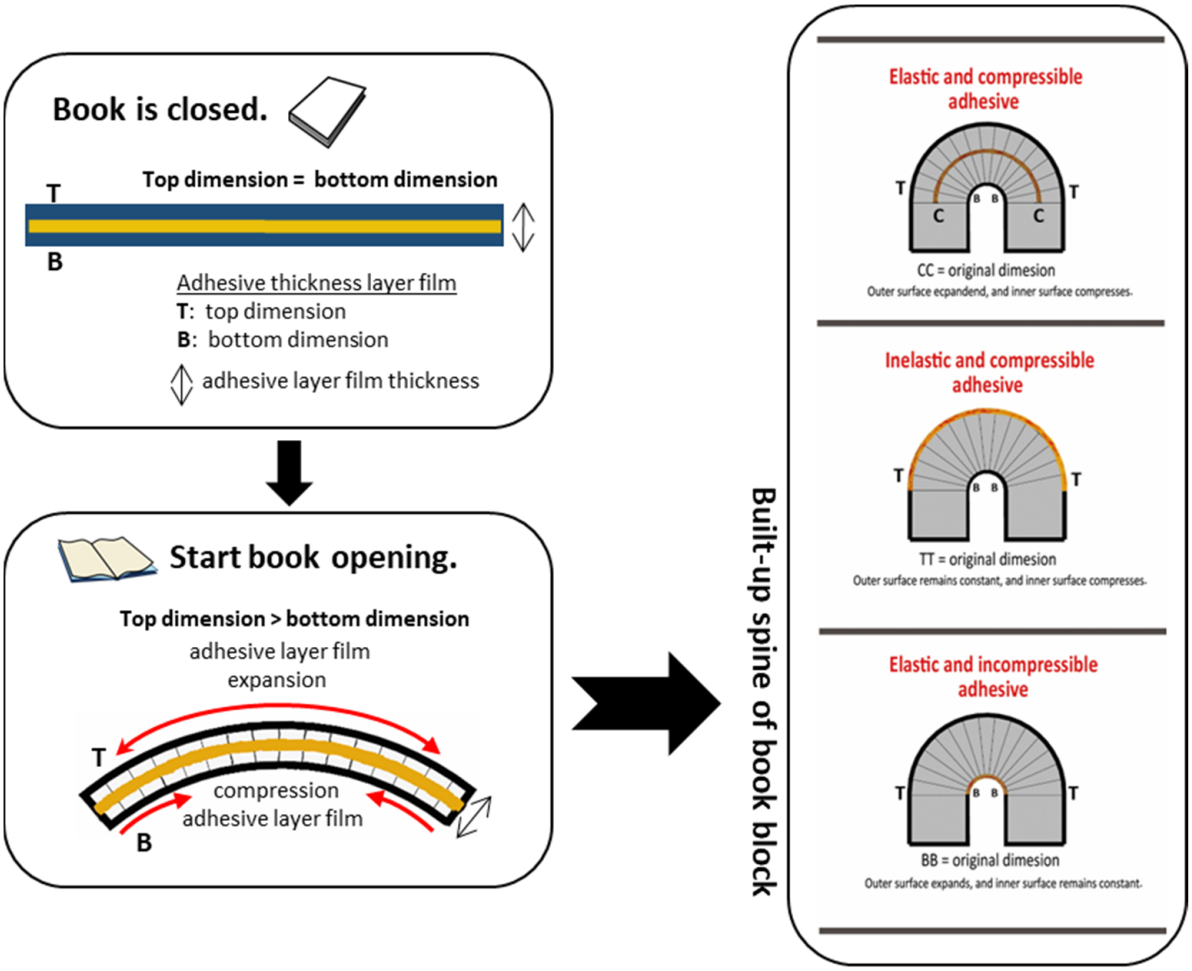

Figure 2. The build-up of the book block spine structures.

A useful adhesive in the binding style must move with a book block spine. When the book opens, the adhesive will crack if it is inflexible. In bindery, inflexible or stiff adhesives are thermoplastic resins of polyethylene vinyl acetate polymer (the EVA hot melt adhesive), animal, and paste adhesives. The reactive thermoplastic resin polyurethan (PUR hot melt adhesive) and the PVAc adhesive are the flexible ones. The inflexible adhesives tend to control the movements of the book block spine more than flexible ones. By increasing the adhesive layer thickness, spine control increases and the stiff adhesive is not able to accommodate its dimensional changes or returns to its initial shape. Therefore, the EVA hot melt adhesive is ideal for unsewn and thread-sewn paperbacks. Generally, binders prefer more flexible adhesives that can provide flat book opening (hands-free reading) even with heavier paper stocks. In addition, the ensuring of bound books' quality in rank from low to high page counts and the adhesion of different printed substrates into book block is important as well. Their strength and durability are excellent choices for books that need to withstand a lot of handling, such as magazines, yearbooks, catalogues, manuals, children's books, etc. The reactive PUR hot melt adhesive, in perfect edition binding, binds to fibers at molecular level, which is a key factor in its durability with respect to widely ranged paper grades, while the PVAc adhesive is mostly used in binding styles, such as Otabind, Swiss, and Double fun. As previously mentioned, the EVA hot melt adhesive can

also improve better book-open control with a higher thickness of adhesive film layer on the paper substrate.

Ethylene vinyl acetate is a rigid synthetic resin material which has a large scope of application in edition binding (education books, belle letters, color books, etc.) including different office and bulky uncoated paper grades. The rough paper surface on a microscopy scale ensures that the polymer adhesive will adhere to crevices on paper surfaces. Thus, they are held together by mechanical forces, in which the thickness of the adhesive film layer defines bond strength through the square-backed spine of the paperback. The EVA hot melt adhesive ensures better adhering results when plasticizers are integrated, resulting in reduced adhesive stiffness. Likewise, the same procedures should be performed with the PVAc adhesive. In general, PVAc adhesive is a liquid substance, diluted with water or with a starch (paste) adhesive. When completely dried, it cannot be diluted in water. Library and craft binders use this adhesive in all binding operations (Figure 3) because of its humidity and heat resistance, and because of its ability to maintain its perfect elasticity without aging problems such as bacterial and fungicidal attacks over considerable storage time [13]. Adhesive plasticizers mostly contribute to the higher bound strength of bound products, ensuring lower creep dimensional changes under load and excellent water resistance. However, they create difficulties during equipment cleaning in handbook binding. A relatively higher amount of non-adhesive substance can reduce adhesive penetration into substrates. Another option is the usage of a paste (glue) adhesive in library and handbook binding operations (Figure 3). They are prepared by the heating of starch and water. However, the growth of microorganisms always causes the weakening of the adhesive bonds and the development of foul odors. In terms of end-product quantity, the unsewn binding style is the most important and the most widely used production process of books in bindery. Therefore, PVAc adhesive, EVA hot melt, and reactive PUR hot melt adhesives must improve the adhesion on various paper grades surfaces [35]. The choice of adhesive system solution depends on the choice of materials to be bonded and on the existing machinery in the bindery sector.

Design and Environmental method / **Adhesives binding options**	PUR hotmelts	EVA hotmelts	PVAc emulsion	Animals	PUR hotmelts	EVA hotmelts	PVAc emulsion	Animals	EVA hotmelts	PVAc emulsion	PASTE emulsion	PASTE + PVAc	PVAc emulsion	PASTE emulsion	PASTE + PVAc	PVAc emulsion	PASTE emulsion	PASTE + PVAc
Binding processes	Long-run production ISO 16763, ISO TC-130				Middle-run production ISO 16763, ISO TC-130				Short-run production ISO 16763, ISO TC-130				Hand binding operations ISO 16763 ISO TC-130			Library binding LBI Z39.78-2000		
Perfect (Adhesive) Binding style	+	+			+	+			+	+			+			+		
Finland Otabind Binding style			+				+			+			+			+		
Swiss Binding style			+				+			+			+			+		
Double-fan Binding style (attaching the Loose Leaves in Book Block)			+				+			+			+			+		
Case making				+			+	+		+			+			+		
Case-in			+				+			+		+	+	+	+	+		
gluing Endpapers			+				+			+			+		+	+		
gluing Sawn Spine of Book Block	+	+			+	+			+	+			+		+	+		
lining the Spine of Book Block (mull, stiff boards, crepe paper, kraft paper, liner)			+				+			+		+	+		+	+		
Adhesive using frequency	2	2	6	1	2	2	7	1	1	9	0	2	9	1	4	9	0	0

Hard recycling waste	Partly hard recycling waste	Recycling waste	Neglecting waste

Figure 3. Environmental comparison of adhesive systems' impact on different binding options.

Generally, machines working with dispersion adhesives are limited due to processes which require a certain time period for water to evaporate (Figure 3). However, hot melts enable high speed production (Figure 3) by application of adhesives via rollers, requiring less equipment maintenance and energy consumption. Reactive PUR hot melt adhesives provide a high adhesion capacity on various paper grades edges. They are resistant to aging and provide higher machine speed book production per hour. However, reactive PUR hot melt adhesives produce VOC emissions, which are categorized as hazardous substances (Figure 3). Undoubtedly, the adhesive joint designs of the EVA hot melt adhesive, the PUR hot melt adhesive, or the PVAc adhesive systems obtain different mechanical performances of binding quality on various adherend substrates. Hot melt adhesive systems (the EVA hot melt and the PUR hot melt adhesive) offer sensitive thermoplastic resins, which can easily be damaged by heat. Thus, the bonds become brittle at lower temperatures, while increasing the temperature causes higher resin softening levels [36]. Therefore, the joints need to be designed with the proper adhesive setting and minimum stress concentration. Thus, the adhesive and adherend material choice and their joint geometry perform the degree of interfacial contact [37,38]. Above all, adhesively bound joint system performances must be considered to improve the strength and useful life of the end-product. In bindery, the most widely used is the PUR hot melt adhesive that ensures durable and long-term performance in edition binding, while the leading place in library and handbook binding is the PVAc adhesive system. These adhesive systems have a maximum percentage of the bound areas and on various paper grades (recycled, office, bulky), performing minimized adhesive stress. These adhesive systems are weak and peeling under tensile loads because the adherends and adhesives tend to stretch. Graphic arts materials in bindery directly perform stress distribution due to the specific mechanical (tensile stiffness) and rheological (viscoelasticity) properties of materials [39,40], which tend to bend under loads. Therefore, the carefully controlled storage conditions (T = 23 °C, RH = 50%) of materials ensured only certified adherend substrates and adhesive systems (ISO 16763, ISO/TC 130, LBIZ39.78-2000) [15,17,18] since the physical properties of materials are affected by its moisture content. Working with adhesive systems must prevent the harmful effect of hot melts through the adhesive safety data sheet, which contains details about hazard rating (exposure control), its handling, and storage. It is obligatory that binders develop a written hazard communication program in which they will provide information and training of employees about the hazardous chemicals found in their workplace. In the bindery sector, all participants (suppliers and process control stakeholders) must be included in multiple graphic arts processes (Figure 3).

3.2. The Eco-Efficiency Guidelines for Adhesive Applications in Bookbinding

There is a wide range of substrate materials which can be used in book binding, so different adhesive systems are being developed to satisfy these needs, especially in edition binding (Figure 3). Various paper adherend surfaces must be wetted with a "liquid" adhesive (cold water emulsion and hot melted resins) which make bonds too hard and permanent. A bond strength is maximized when the solidification of bonds is complete (the closed adhesive dry time) and always correlates with the adhesive molecular mass of used synthetic or natural polymers. Hence, adhesive cohesiveness rises with the higher molecular mass that reduces the adhesive-wetting ability on adherends. The applied adhesive systems in edition binding require low costs which contribute to high processing speed, including fast setting times, especially with thermoplastic polymers. Their wet tack refers to the initial cohesive strength of the adhesive film layer before solidification, which is always higher than that of natural polymers. Thus, thermoplastic polymers can hold two adherend surfaces together after they have been pressed in contact before adhesive setting and its solidification. In library and handbook binding, in contrast, the water-based adhesive systems cannot perform the initial cohesive strength because of the lower molecular mass of the polymers. In this situation, the water-based adhesive system develops bond strength only by pressure. In addition, the water in the adhesive

does not resist motion as a result of its higher attractive force with cellulose fibers on the adherend surface. The water is a carrier of polymer resin. When the evaporation of water from the adhesive ends, the adhesive solidification finishes on the paper surface and the polymer film layer is formed on the surface of the paper. Figure 3 presents a specific bindery method in environmental work conditions, which directly contributes to eco-efficiency and sustainable binding.

The DfE method [9] ensures optimal inputs of adhesive systems, energy consumption, and reduced emissions in the bindery sector. The DfE method is closely related to sustainable binding production, with up to-date information on the environment and the binding style production knowledge. It is tailored to binding production participants as mentioned in Framework for ISO/TC 130 Standards [17]. Within this procedure, it is possible to track environmental indicators and share the information with other suppliers and stakeholders in the graphic arts industry.

This qualitative DfE methodology includes action research and case studies in order to illustrate real-life binding situations, according to the environmental aspects and standardized procedures described in ISO 14005:2019- Standard [41]. DfE method [42] reduces the environmental impact of an end-product that is compared to the estimated capacity of the Earth, regulated in capacity by the World Business Council for Sustainable Development (WBCSD) [43]. The environment indicators present intensity ratios which calculate the quotient of the environment aspects of the binding loads recourses and their utilities. As previously mentioned, the paperback (magazines) edition binding with the PUR hot melt adhesive system in mass production has a significant impact on the Earth's carrying capacity. Hence, sustainable development is needed without compromise. This is the direct value indicator of each participant in the bindery sector through financial, production, or service utilities, including economic, social, and environmental areas of interest. Generally, the DfE method provides sustainable approaches in which non-renewable adhesive systems should be easily detected. Thus, the designed construction of paperbacks, with the appropriate adhesive system, has an optimized life span. It means that non-renewable adhesive systems, with high molecular mass need to have remanufacturing possibilities. Therefore, binders should replace them with "eco-friendly" adhesive systems. Advanced adhesive systems knowledge and up-to-date information together with novel approaches (biodegradable and compostable hot melt biomaterials) and enhanced bound binding style solutions should lead to higher level sustainability in the bindery sector.

4. Eco-Efficiency Binding Style Enhancement and Advanced Adhesive System Solutions

In the bindery sector, maintaining the consistency of adhesive systems is a crucial factor. It means that process and material management should present the best results within the manufacturing processes. Therefore, the binders and suppliers together must seek and develop novel sustainable adhesive system solutions with strong adhesion on a variety of substrates. Novel formulated adhesive systems should exceed international standards and continually ensure technical support to participants in the graphic arts industry.

Above all, novel eco-labeled (non-toxic) adhesive systems should provide the optimal quality standards in manufacturing speeds and the broadest temperatures range. Advanced adhesive system solutions should be focused on hot melt adhesive system performances and their reduced production cost, which should significantly improve binding operational processes such as page pull-flex stability tests, aging, and bond strength line stability, including different ink and varnish systems, in digital technologies. High performance of eco-labeled adhesive systems should be suitable for a wide range of applications, including hardback and paperback binding editions on demand. These eco-labeled systems should achieve an optimum adhesive performance in various "green chemistry" types for different end-products. Sustainability in the eco-certified bookbinding value chain means inclusion of a circular economy principles and life cycle assessment methods (LCA) [44]. In this

circular loop, the graphic arts materials can be returned into the circular loop as "waste back turning into valuable resources". The whole procedure should be obligatory for each participant included in the binding processes. Therefore, novel sustainable approaches to book designing concepts with eco-certified graphic arts materials under standardized procedures encourage responsible business and leads to zero pollution. This analytical tool promotes a new developing perspective of enterprise [45–48]. There is comprehensive literature dealing with the experimental behavior of advanced adhesive system solutions. "The green technologies" include various natural raw materials which can be added into the adhesives formulation. Resulting advanced adhesives are non-toxic and recyclable. Eco-advanced adhesive systems are very important in mass perfect binding production. Gluing procedures are always executed with thermoplastic synthetic raisins including reversible EVA hot melts or irreversible reactive PUR hot melts, which are derived from petrochemicals. The converting adhesive quality must be determined throughout standardized procedures (ISO 16762, ISO 16759, ISO 16763, ISO 20690, ISO 21632) [18,49–52] and some results of adhesive systems quality determinations have been comprehensively presented in the researched literature [35,53–63]. Due to petroleum resources, in current hot melt adhesive system solutions, the return of raw materials into the circular loop cannot be achieved through LCA Management support. Therefore, the adhesive component that makes up the final bound-book product should be standardized, compostable, and/or recyclable by employing standardized eco-labeled paper grades [64,65]. In addition to this, the performances of the new eco-labeled adhesive solutions must establish the full attachment to exposed paper fibers on a book block spine avoiding paper cohesive failure, in which new eco-adhesive formulae lead to the performance of favorable bonds. The work of adhesion through the development of interfacial bonds must improve durable and strong adhesive joint performance [40]. Correspondingly, favorable mechanical properties of adherends could be reached by residual internal stress, favorable interfacial contact degrees, and joint geometry. The page pull-flex stability is realized through reducing stress concentrations, when the loaded stress passes across the whole bound area of book block spine (Figure 2). The adherends which are inflexible under loading often end up with splitting the adherends apart. From a sustainable point of view, advanced adhesive solutions should achieve standardized reliability and repeatability of adhesive bonds, exclusively by using certified paper grade substrates [38].

Conducted studies showed that hot melts (petrochemical-based resins) can be replaced by biomaterials [61,66–78]. New renewable thermoplastic adhesives, which are non-toxic and biodegradable, are developed by groups of researchers. These advanced adhesive system "hot melts" solution offers excellent hot tack with the combination of long open dry time and moderate time settings. Unfortunately, the bond strength and durability of "eco-hot melts" system solution needs to be achieved in further research [66]. Heinrich (2019) gave a critical overview on the advantages of bio-based polymers, including their novel functional molecular architectures which improve adhesive curing speed and bond strength [61]. New bio-based polyurethane adhesives with different biomaterials (biopoly oils, palm oil, soybean oils, vegetable oils) are presented as less expensive and more available on the market [66]. Moreover, this advanced PUR adhesive solution improves further competitive performance such as bonding strength, water and peel resistance, adhesiveness. and thermal stability. Finally, Magalhaes et all. (2019) found out that the large macromolecules of biopolymers, with a high density of functional groups, should lead to higher crosslinking densities [67]. Thanks to their molecular structure, society is going to be less dependent upon the petrochemical resources; hence, the carbon footprint will decrease. These sustainable engineered "hot melts" adhesive system solutions tend to be renewable and recyclable. Generally, advanced "hot melts" technologies ensure health and safe environments supporting the circular economy [68]. Comparatively, a group of Asian researchers [69] studied the bio-plasticizers that should replace the current phthalate-based plasticizers. Traditional phthalate plasticizers in the EVA hot melt systems and the PVAc emulsion adhesives [70] are most compatibles with resins. They modify ad-

hesive rheological properties improving adhesive flexibility. New components in adhesives (non-toxic plasticizers) are derived from biomass sources and agricultural waste such as starch and cellulose. Additionally, in their research, Kim et all. (2020) [71] confirmed that the higher degree of saponified polyvinyl acetate adhesive in hot melt adhesive provides its faster biodegradation under natural conditions. The novel modified polyvinyl acetate emulsions are grafted on biopolymer-based emulsion such as starch, cellulose. and lignin. Gadhave and Dhawale (2022) found out that the grafted vinyl acetate polymerization of corn starch provides adhesive thermal stability and bonding strength [72,73]. Undoubtedly, the grifted polymerized vinyl acetate from chitosan provides excellent bonding of the chitosan-graft-PVAc adhesive, which is an environmentally friendly adhesive. Finally, starch-based adhesive modified by cross-linked starch is a natural polymer is renewable and biodegradable. Novel studies show that starch ethers and esters could be used as components in hot melt adhesives. These components occur at the lower melting points during which adhesive becomes less harmful [74]. Another one, high-performance synthesized lignin biobased adhesive, could find potential application in library binding due to its chemical stability towards acidic and basic media with different adherends [75,76]. Furthermore, environmental benefits of polysaccharidic adhesives, which are derived from organisms (animals, plant, microorganisms), offer a large choice of macromolecules which could provide adhesive performance in the research that follows [77]. In addition to the previously mentioned, the possibilities of eggshell waste utilization could provide additional potential in research work due to the eggshell filler's bonding capacity [78].

In addition to the performances that must be met in order to ensure additional sustainability and good adhesion, all stakeholders included in the graphic arts sector should also explore and investigate some other problems related to adhesives and papers: namely, problems that appear in paper recycling processes. One big problem related to adhesives in the graphic arts industry are stickies, a large particle from various mixtures of synthetic polymeric organic materials occurring during the paper recycling process. Regardless to the fact that most of the printed books will not be recycled after a short lifetime, with regard to the precautionary rule, the behavior of the adhesives used in bookbinding should also be taken into account in the paper recycling process. This relies on the fact that after printing and bookbinding, the production of scrap containing glued paper occurs. Those rejects may end in the paper for recycling. On the other hand, some magazines, instead of staples use adhesives for binding, which again found their way to paper recycling facilities. The paper recycling process aims to convert recovered paper to new products or to recover the cellulose fibers. In most cases, recovered paper or paper for recycling is printed, glued, coated, etc. All the present substances that are not cellulosic fibers are not desirable in the final product and should be removed at the highest degree possible. Deinking by flotation is the most common paper recycling practice. It is often supported by different chemicals used for the improvement of recycled pulp optical properties [33]. However, this process is still not good enough for the removal of stickies. Stickies can create specks, i.e., holes in the final product and lead to process disturbance. In general, all components that can create a sufficient adhesion and cohesion can be a source of sticky impurities. Moreover, stickies have to be liquid, or soft enough at higher temperatures to form sufficient adhesion bonds, and must be big enough and have enough cohesion to achieve noticeable effects. In most cases, pressure sensitive adhesives (PSAs) show sticky potential. In the last decade, manufacturers have started making adhesives that are easier to remove during paper recycling removed by slotted screens and flotation. These adhesives are based on hot melt adhesives and called recycling compatible adhesives (RCAs). In order to achieve better sustainability of the processes, those hot melts are suggested to be bio based.

Novel Advanced Adhesive System Solutions in Bookbinding (Modified Nano PVAc Adhesives)

A Spanish group of researchers [79] found that nanoparticles can be ecotoxic because they can damage the organisms' cell structures. In order to understand nanotechnologies better, the researchers described and classified nanoparticles according to their life cycle.

These nanometer scale particles generate a wide range of applications due to their different properties and characteristics. Their environmental impact is not always negative, but it is very important to know and to understand how nanoparticles are incorporated into the end-products and how they are utilized. Establishing protocols through detailed legislation should ensure tracking uncontrolled variables during fabrication, disposal, and recycling of nanoparticles. New green nanotechnologies provide alternative ways to produce relatively pure nanoparticles, which have low toxic and high biodegradable characteristics. Their environmentally positive impact directly promotes advanced production systems including renewable energy sources (solar, wind, geothermal energy collecting) and remediation of water and soil, which minimize CO_2 emissions that come from fossil fuels. A Canadian scientific group of researchers [80] incorporated nanoparticles into emulsion-based adhesives. Their shapes and sizes affect the enhancement of mechanical, thermal, barrier, and optical properties of adhesives. Moreover, proper dispersion of nanoparticles in polymer matrices is very important in achieving adhesive performance. Indeed, nanoparticles have highly specific surfaces; hence they tend to agglomerate and create heterogeneous dispersion. Petković et al. (2019) [81] incorporated different concentrations of silica (SiO_2) and titanium dioxide (TiO_2) nanoparticles into water-based emulsion polyvinyl acetate adhesive. They found out which is the optimal nanoparticle concentration in advanced nano-PVAc adhesives in order to achieve good adhesive properties. The PVAc-paper adhesive joint's quality was determined to be a highly complex issue because of specific paper grade properties [82]. Conducted research has shown that the modified nano-SiO_2-PVAc adhesive improves physical bound-book performance during handling (opening behavior) for paper grades that resist bending [83]. In comparison, the modified nano-TiO_2-PVAc adhesive was significantly more resistant to temperature and moisture changes than the nano-SiO_2–PVAc adhesive [84,85]. For both modified nano-adhesives, the incorporation of nanoparticles (SiO_2 and TiO_2) did not make significant changes to the color. It was shown that modified nano-adhesives form an adhesive layer that is suitable for end-products manufacture, with a visible adhesive line [86].

In the past, adhesives have played a significant role for many different applications. These materials have been designed, engineered, and manufactured as petroleum based, long lasting, and one use only, i.e., not designed for reuse or recycling. A move towards more green adhesives derived from renewable resources which use less or nontoxic recourses, low VOCs, low energy, and minimizes waste, both during manufacture and at EOL, is a positive step forward. Recently, adhesives are being designed as renewable or biobased derivatives; for example, carbohydrates, tree resins, lignin's, unsaturated oils, and proteins. The traditionally linear petroleum-based economy in the production of different materials is transferring to a circular bioeconomy, with a sustainable approach in the production and use of chemicals, materials, and energy. In addition, the production of books should include green chemistry principals as well. Design of new processes and products should be approached from the sustainable perspective, incorporating the circular economy and green chemistry [87].

5. Sustainability in Library Binding

Global green initiatives in the publishing industry are increasingly implemented with the aim of reducing the carbon footprint created by the production, disposal, and recycling of bookbinding waste. Such programs directly promote the production of green books that are printed on certified recyclable papers. Unfortunately, we are witnessing the fact that conventional adhesives are still used in bookbinding due to their efficient performance, although participants show concern for the environment. Fortunately, there are environmentally friendly initiatives for them as well, and their new sustainable solutions meet the performance requirements for various bonding technologies. It is important to emphasize that bookbinding should be viewed from the aspect of cultural heritage, which is not sustainable without knowledge of the bookbinding craft. Therefore, sustainable development with raised the standards and quality of execution with respect to the bind-

ing concept must be standardized in accordance with the purpose of the book. When the norm is mentioned in the context of measuring the worker's performance, then the norm is determined by several elements: a certain amount of product, rational use of means of production, a certain time, and certain degree of technical and organizational working conditions.

In library binding, the primary goal is to prolong the existence of the book as a cultural good. Moreover, the main differences between library and publishing bookbinding lie in the used processes and materials, which arise from the purposes of the bonded books. Library binding is a hand work process, while machines are used in publishing binding. In library binding, when one or few books are being bound, the achievement of good strength properties in the end product it important. In contrast, in publishing binding, the mass production of large book volumes is required often results in low bonding quality. In the case of library binding, it is often mentioned that it is important to know the materials (mainly paper and covers materials), methods, finish, and use. Librarians who deal with binding should take care of materials and processes used in the binding process. The process should meet not only conservation standards but technical standards as well, in order to obtain a high-quality and long-lasting product [15,17]. Materials should be sustainable, less waste should be produced during binding process, there should be less use of harmful and toxic chemicals, etc. This paper points to better knowledge in bookbinding processes in publishing binding but also the importance of the implementation of these technologies in library binding as well.

Books as cultural goods should be preserved for future generations, in accordance with sustainable development goals, since they are key factors in the community's social development. However, books are vulnerable cultural goods due to environmental changes occurring in their surroundings. Different gases, humidity, and temperature can affect the mechanical properties of the books. In order to prevent this, it is important to make small changes in all segments of life in general and all branches have to take into consideration their environmental impact. Librarian bookbinding is one of them. Although library binding, considering the few editions of the books that are bound, is less harmful to the environment in contrast to publishing binding, it is important to think locally in all segments in order to achieve global sustainability goals, i.e., the goals of environmental protection. This can be achieved by the development of different policy documents offering analysis, information, and education to all stakeholders about the issues and solutions regarding the occurring problems.

In library binding, the protection of cultural goods should be conducted by activities that include: proper care and handling by the controlling of environmental temperature and humidity, creation of enclosures, security of the facility and the collections, physical or chemical stabilization, and digital preservation [88].

The research conducted by Rachman and Ratnasari [89] shows that current practices of sustainable library preservation and conservation implemented in the majority of academic libraries relate to sustainable energy use. In addition, the research showed that the majority of libraries included in the research demonstrated good environmental awareness and implementation of sustainable practices in library preservation and conservation. Moreover, the research showed the need for regulation toward sustainable library preservation and conservation practices, alternative eco-friendly preservation and conservation materials, and waste management practices.

Most of the studies related to the library conservation and preservation are related to the causes of material deterioration, elements of conservation, prevention measures, and implementation of digital preservation. The lack of research related to sustainability in bookbinding (development of novel eco-friendly adhesives with minimal costs) suggests that this could be a promising research field in the near future.

6. Conclusions

The main goal of this paper is to provide a comprehensive review of adhesive applications in the bindery sector. The manufacture of bound books requires sophisticated adhesive system solutions for very common products such as paperbacks and hardbacks. Certain book binding styles must provide specific combinations of adhesive strength capacity, elasticity, solidification, and usability on a wide variety of paper grade edges. Undoubtedly, the different printing possibilities with respect to traditional and digital technologies on paper substrates leads to the limitation of adhesive applications. Unfortunately, in modern bookbinding, adhesives are derived only from a few groups such as emulsion solutions, hot melts, or reactive hot melts. Moreover, conventional adhesive formulations should be advanced in a way that novel modified adhesive system solutions contribute to better eco-efficiency, especially in perfect adhesive binding, which includes the "one-shot adhesive process" in long-run paperback production. The novel sustainable adhesive system solution concepts enhance adhesion on a variety of substates and connect all stakeholders that participate in the development of novel eco-labeled adhesives for Graphic arts industry. Whereas adhesive hot melt system solutions are dominant in edition manufacturing, the emulsions are dominant in hand bookbinding, including librarians. Nowadays, worldwide, scientists tend to offer sustainable "green adhesive" technologies from various natural sources which are being incorporated into adhesive formulations. These superior advanced adhesive system solutions provide increased manufacturing performance, both for bookbinders and librarians. These sustainable sources, which are incorporated into adhesive systems, will soon ensure their certification and traceability according to environmental management standards (Life Cycle Assessment and Environmental Management). Another step should be the development of a framework of sustainable workflows models in the bindery sector, which will serve as a tool for the identification of opportunities in sustainability including environmental, social, and economic aspects. A good example is circular resources management (graphic arts materials and semi-products) via the closed-loop and useful lifetime of the end-products (repairing, re-using, re-manufacturing, recycling). This is a new way of connecting suppliers and customers (closed-loop manufacturing) in which a new form of partnerships is supported. The new sustainable business model includes circular suppliers, resource recovery, product life extension, development of shared platforms (social networks). and novel products through advanced services. The established "green models" should provide sustainability since each participant contributes to the closed-loop productions within the graphic arts industry.

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