# A review of recent developments of water and energy optimisation methods applied to Kraft pulp and paper mills

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# ABSTRACT

This review presents recent developments of water and energy optimisation methods applied to Kraft pulp and paper mills. The Kraft process has the potential to be adapted to biorefineries for producing biofuels and other high-value products from wood biomass. It increases the revenue of the process, reduces fossil fuels usage and greenhouse gas emissions. The results of several case studies of the Kraft process are presented to show typical savings in terms of reduction of water and energy consumption by applying systematic methods described in this review. In these case studies, freshwater consumption is decreased by between 20% and 81% and energy consumption between 15% and 40%. Recent studies show that research fields related to water and energy optimisation in pulp and paper mills and Kraft process-based biorefineries have attracted attention from researchers in the last several decades and will continue to be the subject of further research.

# **KEYWORDS**

Water and energy, systematic methods, pulp and paper mill, Kraft process, biorefinery.

# INTRODUCTION

Pulp and paper mills (P&PM) are large consumers of water and energy and emitters of greenhouse gases (GHG) [1]. By improving energy integration in P&PM, and using steam and electricity produced from black liquor, the use of fossil-based energy and GHG emissions can be significantly reduced. If steam and electricity obtained from black liquor are used for all process energy requirements, recycling of water is maximised and wastewater discharge to the environment is minimised then significant environmental and economic benefits can be

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obtained. As a result of synergistic interactions between energy and water consumption, reducing one consumption usually causes a reduction of the other. Accordingly, systematically and simultaneously exploring water and energy interactions between P&PM and utility systems is important for minimising overall water and energy consumption, wastewater generation and harmful gases emissions to the environment.

Process integration and systematic methods based on pinch analysis (PA), mathematical programming (MP) or their combination have been used for minimising heat and/or water consumption in the process industries. These methods have been used for more than four decades for heat integration [2], and more than two decades for simultaneous optimisation of water and heat integration [3]. Klemeš and Kravanja [2] present a review of PA and MP methods for heat integration and opportunities for employing a combined PA/MP approach in heat integration. Ahmetović et al. [3] present a comprehensive literature review of concepts, methods and solution strategies for the synthesis of non-isothermal water networks and performing combined water and heat integration by using PA, MP, and their combination (PA/MP). A recent paper by Kermani et al. [4] provide a review of methodologies with special focus on MP approaches, their key features and solution strategies. Different concepts, methods, and solution strategies have been developed and applied for exploring combined mass/water and energy/heat integration opportunities in various processes. A recent review [3] presents industrial case studies in which the concept of non-isothermal water networks is applied, and case studies in which other concepts are used for water and energy integration in various processes, including P&PM. A review paper on process integration in P&PM for energy and water reduction is given by Atkins et al. [5]. This paper focuses on the analysis of different techniques for separate heat integration and mass/water integration. A brief review of several works is given for simultaneous heat and mass/water integration. A review provided by Mahmoudkhani and Berntsson [6] highlights that there is a lack of works which present a summary of various methods and tools for process integration and their application to P&PM. This review presents methodologies developed and applied in process integration studies for Kraft P&PM in Sweden. The major focus is on energy integration, but it does not present a review of studies related to combined water and energy integration performed by MP in P&PM. Combined water and energy integration has been an active research area in the last two decades, and the proposed optimisation methods have been successfully applied for water and energy reduction in P&PM. To the best of our knowledge, there is a gap in up-todate literature review of papers in which various concepts, methods and solution strategies are applied for combined water and energy integration in P&PM.

The goal of this study is to present a review of recent developments in optimisation methods and solution strategies for water and energy integration, which are applied to Kraft P&PM. The paper is organised as follows. Firstly, a brief description of P&PM including, an overview of water, energy, and material flows, is given. Then, a review of systematic methods for water and energy integration in P&PM is presented, including the results of case studies to show typical savings in water and energy consumption. Finally, conclusions and possible future directions in this field are highlighted.

## **DESCRIPTION OF PULP AND PAPER MILLS**

An integrated P&PM comprises two production processes. In the first, wood is converted to pulp, while in the second one pulp is used for producing paper products. There are different methods (chemical, semi-chemical, chemi-mechanical, and mechanical) of pulping processes in which pulp is obtained from wood [7]. A well-known and commonly used process for producing wood pulp and high strength paper is the Kraft (sulphate) process [8]. Large

amounts of the pulp produced throughout the world are obtained with this process. In a typical Kraft process, wood logs are debarked and chipped for de-lignification process in a digester. A white liquor consisting of sodium hydroxide (NaOH) and sodium sulphide (Na<sub>2</sub>S) is used for cooking the wood chips in the digester and partial deconstruction of biomass. The lignin and part of the hemicellulose are dissolved in the white liquor. The pulp obtained is washed, bleached, and provides the main cellulose component for producing paper products. The resulting main by-product of the Kraft pulping is a cooking liquor called the black liquor, consisting of degradation products of the lignin and polysaccharides, with a minor fraction of extractives [9]. After separation of the weak black liquor from the pulp, this liquor is concentrated in an evaporation plant, which is an energy-intensive process. Multiple-effect evaporation plants usually consist of four to eight evaporators is used for improving the steam economy. The strong black liquor is burnt in a recovery boiler to produce steam used for heating and for producing power (electricity), while residual inorganic materials (smelt) from this boiler are reused in the process. Smelt, consisting of sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>) and sodium sulphide (Na<sub>2</sub>S), is dissolved in a tank with the weak liquor. The green liquor produced in the smelt dissolving tank is recaustified with lime to regenerate the white liquor reused in the digester. More details about the Kraft P&PM process is given by Bajpai [10] and Särkkä et al. [11].

Figure 1 shows an overview of water, energy, and material flows in a P&PM. Understanding the interconnections within a P&PM is very important for reducing water and energy consumption. A complete system consists of water preparation, utility systems (steam and power plant and cooling tower), a P&PM integrated with biorefinery technologies and wastewater treatment. This figure shows closed loops for steam and condensates, cooling water and warm water, process water and wastewater. There are also two connecting loops of sodium and calcium in a pulp mill for recovery and reuse of chemicals (NaOH and Na<sub>2</sub>S). Raw water is purified in a water preparation system which is used for different purposes in the system. Some of this water is directed to boilers (biomass boiler, coal boiler and recovery boiler). Different fuels are burned in the boilers to generate heat for heating and evaporating water and producing high-pressure steam. Fuel for a biomass boiler is bark and hog fuel. The biomass fuel is generated in the wood preparation system of the P&PM (debarking and chipping process). In the second boiler, fossil fuel is coal and in the third, biomass fuel is the black liquor coming from the P&PM. Flue gasses and solid materials are generated in boilers by burning bark, hog fuel, and black liquor. Carbon dioxide emissions from the coal boiler are usually associated with GHG emissions, while the combustion of the black liquor, hog fuel or other biomass sources are treated as carbon neutral and not usually counted in GHG emissions [12]. Smelt from the recovery boiler is used for chemical recovery in the sodium and the calcium loops. Blow-down from boilers and cooling tower including wastewater from the process is sent to the wastewater treatment plant. Part of the water from wastewater treatment can be reused/recycled in the process, if this water is of appropriate quality. Effluent is discharged from wastewater treatment plant to recipients.



Figure 1. Overview of water, energy, and material flows in pulp and paper mills.

Steam (usually high-pressure) from boilers is directed to a steam turbine. Electricity is produced in the steam turbine, and used in the overall process. When more than enough electricity in the process is produced, some of it can be exported and sold, increasing the revenue of the process. Medium and low-pressure steam is extracted from the turbine for heating in the P&PM. Condensates are collected in the tank, and then sent to the deaerator. Part of the low-pressure steam is usually used for heating in the deaerator. For cooling the hot process streams in the P&PM and condensing the steam after the steam turbine, cooling water is used. Warm water is recycled to the cooling tower to be air cooled. Part of the water is lost by evaporative cooling in the cooling tower.

# WATER AND ENERGY INTEGRATION IN PULP AND PAPER MILLS

Different methods have been applied for water and energy integration in manufacturing processes, including P&PM [3]. Insight-based PA has been used for obtaining water and energy targets before the detailed design of water and energy networks is finalized. The superstructure-based MP method has been applied for sequential and simultaneous optimisation of water and energy integration, including the design of combined water and energy networks. Different studies show that the advantages and synergies of both methods should be exploited in solving complex problems. There are many case studies in the process industries in which systematic methods have been successfully applied for water and energy optimisation. This section presents a review of published works, methods, solution strategies, and results of case studies in P&PM. Typical savings are given in terms of reduction of water and energy consumption.

Early studies in P&PM based on PA and MP focus on separate heat recovery to reduce the amount of external utilities (steam and cooling water) used, and water recovery to reduce

freshwater usage/wastewater discharge [5]. Later works studied the interactions between water and energy and proposed different methodologies for simultaneously reducing freshwater usage, wastewater discharge, and external utilities. One of the first papers related to the simultaneous management of energy and water streams is by Savulescu et al. [13], in which a two-dimensional grid diagram (temperature vs. concentration) is proposed for simultaneous water reuse and heat recovery.

PA presents an essential tool for energy optimisation of P&PM [14]. Process integration tools and the PA method have been widely used in many studies in combination with simulation tools for evaluation of the energy and water savings in P&PM. Simulation tools such as Valmet WinGEMS [15], BALAS [16], and CADSIM Plus [17] can give information about an existing P&PM process and provide necessary initial data for PA. The proposed methodologies for simultaneous energy and water efficiency enhancement in P&PM are usually based on process integration tools, including water-thermal composite curves, the water source and sink curves, and water and thermal PA. These methodologies in most cases follow a project-oriented approach. Savulescu et al. [18] performed a combined energy and water analysis based on PA to reduce water and energy consumption in a Kraft P&PM. Their analysis started with data acquisition and simulation of the mill by the commercial software package WinGEMS to obtain relevant data for performing PA and evaluating potential water and energy-saving projects. They identified nineteen projects, from which twelve were selected after discussion with the mill personnel, resulting in the steam saving of 15% of the boilers' steam production and about 6,000 m<sup>3</sup>/d in water consumption and effluent generation. Also, the effluent temperature is reduced by 3°C by energy and water recovery. To achieve improved heat and water efficiency of P&PM, direct and indirect heat transfers should be considered, including different heat and water efficiency measures (heat integration, water reuse, regeneration and recycling, condensate recovery, and redesign of hot and warm water systems). Direct heat transfer in the Kraft P&PM can include dilution of pulp with hot water, process-process mixing of streams with different temperatures, and process-utility mixing of water and steam. This heat method of transfer can be beneficial with respect to investment cost and simplified designs, but could lead to higher operating costs. Accordingly, an optimal selection of direct and indirect heat transfers in the process should be made to minimise the total costs. Savulescu and Alva-Argaez [19] studied the impacts of non-isothermal mixing of streams and direct heat transfer on the overall energy efficiency of a Kraft process. The importance of data measurement of the streams involved in the mixing when performing a retrofit analysis is highlighted. They proposed a process integration methodology to systematically address the relevant aspects of the direct heat transfer. By implementing of the proposed methodology, the overall energy efficiency is improved by about 4% in their retrofit case study. Mateos-Espejel et al. [20] implemented a five-step strategy to analyse the impact of water reduction on Kraft process energy consumption. Step 1 of the methodology includes water network data extraction. Data used in their study are obtained with computer simulation. Step 2 consists of analysing water reduction potential based on reference mills in operation. Step 3 includes water analysis. Water-thermal composite curves are constructed to identify opportunities for heat integration between hot effluent streams and cold water streams. Water sink and source curves are constructed to explore water integration opportunities and determine minimum water consumption. PA is used to evaluate the energy implications of water reduction on the overall process. Step 4 is used to maximise energy recovery and minimise external utility demand for the defined minimum temperature difference (10 °C). Step 5 presents a trade-off analysis to evaluate the economic indicators of the proposed solutions. In a later paper [21], the authors applied the proposed five-step methodology to the Kraft process and identified four practical strategies for water reutilisation

and their energy implications, by which about 24% of water, 14 MW of steam, and 13.1 MW of cooling demand are saved. Martinez and Picon [22] presented a retrofit concept for water and energy reduction in a P&PM based on the hierarchical methodology and "onion diagram". In the first step of this methodology, the reaction system of the pulp bleaching process for lignin removal is analysed examining trade-offs between additional equipment cost, conversion and water consumption. They found that additional reactor volume gives higher lignin conversion and thus requires less fresh water for dilution reactor outlet stream. The second step of the hierarchy is the water usage system, in which water reuse within the pulp washing stage is maximised by applying a water pinch method. Once the water reuse options are explored, in the third step, wastewater regeneration is considered exploring different distributive wastewater treatment options for each effluent stream and considering equipment cost, operating cost, and practical implementation. The last step includes a heat recovery system. Since the pulping process requires water at different temperatures, a reduction in freshwater consumption consequently reduces heat demand. In this study, only the energy consumption associated with water use is examined. The proposed methodology is implemented using data from a real pulping plant. Mateos-Espejel et al. [23] proposed an unified methodology consisting of five steps (base case definition, pre-benchmarking, interaction analysis, implementation strategy and post-benchmarking), which is applied to Kraft mills to perform thermal energy efficiency improvement. The results show that an ecofriendly process can be obtained without using fossil fuel for steam production, and revenues can be increased based on green electricity produced from biomass. Significant reductions in consumption of steam (26.6%) and fresh water (33.6%) are achieved. This methodology was later extended by Keshtkar et al. [24], including specific features (combined steam and water systems analysis, a project-oriented approach, and use of heuristic rules) and applied to three Canadian Kraft mills with different configurations. This extended step-wise energy efficiency methodology consists of six steps (data base, pre-benchmarking, energy systems efficiency analysis, steam and water efficiency enhancement, retrofit heat exchanger network, and postbenchmarking). The CADSIM Plus simulation software was used in this study to provide all the data necessary to support process analysis. Also, the modified water PA and Aspen Energy Analyser were used in implementing some steps of the proposed methodology. Significant reduction in terms of water (25% to 50%) and energy (30% to 40%) consumption was achieved in this study, compared to current engineering practice, with a payback time period up to three years.

Papers based on MP have focused on optimisation of P&PM to reduce freshwater usage and wastewater discharge, water and energy consumption in P&PM or optimisation of subsystems of P&PM, such as pulp cleaning and black liquid concentration, which are water and energy-intensive processes. MP approaches based on superstructure optimisation including Linear Programming, Mixed Integer Linear Programming (MILP), Mixed Integer Non-Linear Programming (MINLP), have been applied to P&PM to perform optimisation of water and energy integration. Programming languages such as GAMS [25], AMPL [26], and LINGO [27] have been used for modelling. Lovelady et al. [8] presented an integrated approach consisting of process integration (mass integration) and a simulation tool for the optimisation of freshwater usage and wastewater discharge in P&PM. Special care is given to presence and tracing of non-process elements in water streams that can cause process operation problems when streams are recycled. The optimisation program is solved using LINGO software to identify the minimum consumption of fresh water. They reported a 23% reduction in freshwater consumption using simple recycle/reuse strategies, and up to 81% reduction when removal of non-process elements is enabled by introducing technologies for their removal from water streams. This approach considers isothermal water networks, and interactions between water and energy are not explored. Chew et al. [28] explored opportunities for simultaneous water and energy reduction within the brown stock washing system (BSWS) in P&PM by using the MP approach. They proposed a MINLP model to optimise the water network design. This model is applied to BSWS for two scenarios (minimising total operating cost and total annualised cost) and the resulting network is improved in terms of reduction of water and energy consumption (21% and 23%), compared to the base case. The concept of non-isothermal water networks has been recently applied for performing simultaneous optimisation of water and heat integration and the design of these networks in P&PM. Some proposed strategies can generate multiple solutions, from which the best and/or most practical solution can be chosen. Based on this concept, Kermani et al. [29] proposed a superstructure for a combined water and energy network and a step-wise approach, consisting of seven steps (plant characterisation, quantification of qualitative indicators, modelling, preliminary targeting, optimisation problem-solving, identification and evaluation of energy and water reduction opportunities, and project selection and implementation roadmap). The proposed MILP model for simultaneous optimisation of water and energy (SOWE) is used for exploring heat integration opportunities between non-water process streams and water thermal streams. Additional features are incorporated into the model, including restricted matches for water integration, water tanks, and non-isothermal mixing to reduce the number of heat exchangers. The proposed model includes integer-cut constraints for generating multiple solutions, and a heat load distribution model for providing a list of preliminary and promising connections before the detailed design of the heat exchanger network (HEN) is finalized. The objective function of the proposed model minimises total annualised costs, including the operating costs consisting of freshwater consumption, wastewater treatment, utilities consumption and investment cost of heat exchangers. This study is one of the first which attempts to solve a real Kraft process problem using MP, and it proposes promising solutions with improved water and heat integration. A recent work by Ibrić et al. [30] proposes a MINLP formulation of heat-integrated water networks enabling heat integration of water streams with non-water streams (process hot and cold streams) by extending a previously developed superstructure [31]. This model can be used for simultaneous water and heat integration, minimising total annualised costs, including costs of freshwater usage, hot and cold utility usage, heat exchangers and wastewater treatment units. The proposed superstructure includes a water network (WN), a wastewater treatment network (WTN), and a HEN, enabling direct and indirect heat exchange. The overall model is comprised of three sub-models: a WN model (M1), a simultaneous optimisation and heat integration model (M2) [31], and a HEN model (M3) based on stage-wise superstructure [32]. A three-step solution strategy is proposed to facilitate the solution of the overall problem. The first step performs freshwater and utilities targeting by solving combined models (M1+M2). The objective function of the NLP model minimises freshwater and utility consumption and wastewater treatment operating costs. Targets are used as bounds for the following step. The first NLP model is solved for different heat recovery approach temperatures (HRATs) to set the bounds for freshwater and utility consumption. In the second step, models M1 and M3 are combined for simultaneously solving water and heat exchanger networks with the linearised objective function. This step is used to limit the number of matches between hot and cold streams to those existing in the MINLP linearised model. The third step, which combines models M1 and M3, is solved as the second but now minimising non-linear objective function with a limiting number of matches from the second step. The model can be solved in multiple iterations and provides good bounds by changing HRATs. A set of good local optimal solutions can be obtained, from which the best can be selected. The proposed MINLP model [30] is applied to the Kraft pulp mill process [29] by adding restricted connections within the water network due to practical limitations. Cases with no water reuse, maximised water reuse,

and sensitivity analysis related to freshwater and utility costs are presented. Trade-offs between investment and operating costs are established, resulting in a significant reduction in freshwater use (about 34%) compared to the reference case, and 13.5% compared to paper [29]. The total annualised cost of the proposed optimal network decreased 28% compared to the reference case and 14% compared to the optimal solution given in the paper [29]. A recent paper proposed a hybrid MILP superstructure for simultaneous optimisation of heat, water, and power in inter-plant operations [33]. This method is applied to a real industrial Kraft mill and significant reductions in terms of water (about 41%) and low-pressure steam (about 35%) consumption are achieved. This decreases the total cost by about 13% and the operating cost of steam by 22.6% and increases investment cost by 13.8% compared to solutions obtained using traditional approaches (not including the hot water cycle). A recently proposed heatintegrated water allocation network model by Kermani et al. [34] was applied to the same Kraft pulp mill case study. The model is composed of three sub-models: a water and heat targeting MILP model [29], a heat load distribution MILP model for minimising number of matches [35, 36], and a heat-integrated water allocation network synthesis model based on the NLP hyperstructure [37]. Multiple solutions were generated using the proposed strategy. Benchmarking results of the Kraft pulp case study, including key performance indicators, are presented for applied simultaneous [30] and sequential [34] solution strategies, and the results are in a good agreement. However, as can be seen from the proposed optimal design obtained using the sequential strategy [34], the values of some water streams are too small and can be impractical. The benchmarking and key performance indicators for a Kraft pulp case study [34] obtained by applying two optimisation approaches [30, 38] show significant savings. Reductions in freshwater consumption more than 34%, total water consumption (fresh water and cooling water) more than 15%, and total annualised cost more than 27% are obtained. The hot utility is not required in the proposed solutions due to improved heat integration. For this case study, it was also shown [34] that further reduction in freshwater consumption (11%) and total annual cost (< 1%) can be achieved compared to the results reported in the literature [30]. A recent work by Kermani et al. [39] proposes a holistic methodology for optimising industrial resource efficiency. This work considers heat, mass and power integration by combining superstructures for heat-integrated water allocation network [29], and an organic Rankine cycle for waste heat recovery [40]. The proposed MINLP superstructure includes a set of plant sites consisting of water using units and water sources, and a set of hot and cold thermal streams. Mass exchange (water) is managed with water tanks, and heat exchange is managed by heat transfer vector (steam cycle, organic Rankine cycle, heat pump, and water network). A multi-objective framework is considered minimising freshwater usage, maximise power output, and minimise total costs. As trade-offs exist between these objectives, there is no single solution for optimal performance of all objectives; rather, many optimal solutions exist (Pareto solutions). The proposed methodology is applied to an industrial Kraft pulp case study. In this case, a significant reduction in freshwater consumption of more than 60% is achieved, while net power output is increased by a factor up to six (from 3.2 MW to between 10-26 MW). Based on the results presented in this section, we emphasize that systematic methods for water and energy optimisation play an important role in reduction of freshwater and energy consumption in pulp and papers mills.

As shown in Figure 1, P&PM are very complex processes with many interactions between water, energy, and material flows. The advantages of different methods and tools should be combined to solve complex industrial problems. Also, understanding the water, energy, and material interconnections in P&PM, taking into account also practical and feasibility constraints and human interventions at different levels of problem-solving, are very important considerations.

Existing P&PM can be retrofitted and integrated with various biorefinery technologies (hemicellulose extraction, black liquor gasification, lignin precipitation, and tall oil extraction) to satisfy market needs for different products and to increase revenue [41]. Incorporating biorefinery technologies into existing Kraft P&PM, and considering the simultaneous optimisation of water and energy integration of the overall system is very important. Further progress in the development of methods and solution strategies is needed to address this challenge. By properly integrating biorefinery technologies with existing P&PM, water and energy costs and greenhouse gas emissions can be significantly reduced, and different green products produced.

# **CONCLUSIONS AND FUTURE DIRECTIONS**

Future development of methods and solution strategies is needed to improve water and energy integration and their applications to existing Kraft P&PM especially for the purpose converting this process to a multiproduct wood-based biorefinery. Boundaries for water and energy integration should be extended to include combined heat and power system, process system, and water and energy recovery systems. Heat, water, and power integration opportunities should be simultaneously explored, considering synergies between different systems, reducing overall primary energy consumption, and proposing improved solutions to economic and environmental problems. Recent studies show that generalisations for converting existing Kraft P&PM to multiproduct wood-based biorefineries cannot be made, and analysis for each specific pulp and paper process should be performed. The research field related to biorefineries has attracted the attention of many researchers in last several decades and will be an important field for future research. The main goals are to find the optimal alternatives for producing biofuels and other high-value products, improve the water and energy efficiency of the overall process, and to reduce fossil fuels usage, and gas emissions into the environment.

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