# Monitoring of flue gas desulphurization process in coal fired power plants

*Timo Korpela[1], Yrjö Majanne, Tomas Björkqvist*
Tampere University of Technology, Department of Automation Science and Engineering
P.O. BOX 692, FIN-33101 Tampere, Finland, www.tut.fi
[1] Tel: +358 40 849 0063, E-mail: timo.korpela@tut.fi

*Olli Salminen*
Helen Oy, 00090 Helen. Tel. +358 9 617 3573, olli.salminen@helen.fi, www.helen.fi

KEY WORDS Process monitoring, emissions, power plant, diagnostics, sulphur dioxide

## ABSTRACT

The work described in this paper is conducted as a part of Measurement, Monitoring and Environmental Assessment (MMEA) program, which is one of the research programs managed by the CLEEN Ltd. The applications were developed in co-operation with Helen and IndMeas and applied in Salmisaari coal fired power plant. The overall goal of the study is to improve usability and economic efficiency of the desulphurization process by providing the operators assistance and to provide the plant operators early information of arising process problems. The goals are to be met through development of new monitoring applications that assist the plant personnel in day-to-day optimization of the process and help avoiding unexpected shutdowns. This paper introduces an indirect method to monitor calcium slurry spraying in a flue gas desulphurization process. The proposed method to monitor functioning of spraying is based on energy balance, and estimates that are only based on physical calculus were utilized to gain generality. With the method, flue gas exit temperature of the rector, which is the main control variable in reactor control, can be estimated. The calculated temperature estimate indicates what should be the exit temperature if everything were in order. Due to lack of measurements before the sulphur removal processes, several variables had to be estimated based on measurement information and some additional information. Still, the results verity that the method is able to predict the reactor exit temperature by error of typically less than few degrees Celsius regardless of the process state.

## 1 INTRODUCTION

There is an increasing demand to restrict polluting emissions to environment. One harmful emission is sulphur dioxide ($SO_2$), which has unfavourable health and environmental effects. $SO_2$ is formed primarily in combustion of fuels that contain sulphur, i.e. coal and heavy fuel oil. There, the majority of sulphur oxidises, and the amount of $SO_2$ emissions is dependent on the fuel consumption and fuel sulphur content. Currently, the combustion originated $SO_2$ emissions exceed emission standards even with low sulphur coals. Therefore, $SO_2$ reduction methods have been applied since 1980's after the first $SO_2$ emission limits were set for coal fired power plants. Lately, the $SO_2$, $NO_x$ and dust emission limits are further tightened from current Large Combustion Plant (LCP) /1/ directive to Industrial Emission Directive (IED) /2/, which will come into effect in 2016 for existing power plants. According to IED, the $SO_2$ emission limits will be lowered for existing large scale coal fired power plants from 400 mg/Nm$^3$ to 200 mg/Nm$^3$. In Finland, the new emission limits are applied step wisely according to transitional national plan under IED. As the sulphur content of coals is not likely to decrease, the new emission limits are met only with more effective sulphur removal in existing desulphurization processes or with new installations. As investment costs are extensive when building new desulphurization systems, operational improvements in existing systems are extremely beneficial.

There are a few types of sulphur removal processes (SRP). One of them is spray dry absorption, which is applied e.g. in in Salmisaari and Hanasaari power plants of Helen and at Martinlaakso power plant of Vantaa energy. In the spray dry absorption method, slurry constituting of water, calcium hydroxide $Ca(OH)_2$ and recycled reaction products is used for sulphur removal. In the process, the slurry suspension is injected by compressed air through specific nozzles into two parallel reactors, where acid components of the gas, i.e. $SO_2$ and HCl, are rapidly absorbed into the alkaline droplets to form calcium sulphite ($CaSO_3$), sulphate ($CaSO_4$) and calcium chloride ($CaCl_2$) while the water of the slurry vaporizes. With appropriate control of gas distribution, slurry flow rate and droplet size, the droplets are dried by the time they reach the flue gas exit near the bottom of the reactor. Some of the dried products, that contain desirably maximum proportion of end products $CaSO_3$, $CaSO_4$ and $CaCl_2$, minimum amounts of reactive $Ca(OH)_2$ and unreactive calcium carbonite ($CaCO_3$); and water and ash, fall to the bottom of the reactor, while most of the solid particles moves along with the flue gas to bag filters. The filter fabric slowly collects the reaction products, and $SO_2$ removal continues there if the moisture content of the flue gas is at adequate level. Ultra sound and compressed air pulses are used to shake the reaction products to the bottom of the fabric filter units. After that, most of the solid products are recycled to the slurry production system and the rest is discarded as unusable end product. The purified gas then flows out to stack via an exhaust fan. Figure 1 presents a scheme of desulphurization process.

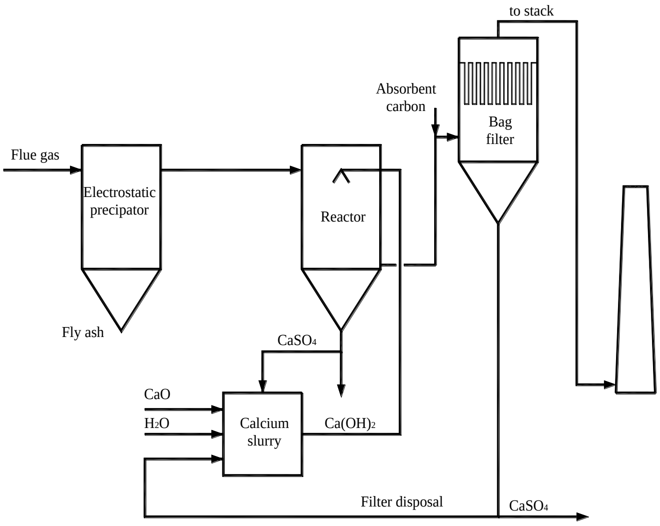

**Figure 1.** Desulphurization process.

The economic background of the method is effective recycling of calcium hydroxide in the process, which causes, unfortunately, several dynamical issues. The challenge with the process is a slow and nonlinear response from manipulated variables to the controlled process state variables. It takes hours before the results of the control actions made to the chemical feeds and flow rates will be seen in the state of the process. Furthermore, the origin of the detected behaviour of the process is often unclear; the reason can be found inside the desulphurization plant or it can be found in the operation of the combustion process as changed flue gas properties. This uncertainty causes easily problems, because the process is sensitive to defective control actions which might lead to unexpected shut downs of the whole system caused e.g. by clogging of the lime slurry lines or spraying nozzles or by overloaded mixers. Therefore, monitoring the operation of flue gas desulphurization process is getting an increased attention. The special concern with process operation is with the slurry injection, where the spray nozzles have a tendency to get clogged up and/or forming obstructions that hinders efficient spraying. In those cases, the droplets are so large that the droplet falls to the bottom of the reactor with several undesired effects, most of which lowered $SO_2$ removal.

The slurry injection is controlled in a way that the moisture content of the flue gas is within limits that the sulphur removal continues at the surface of the bag filters. In practice, this is implemented by controlling the temperature of the flue gas at the bottom of the reactor by slurry flow injection. Unfortunately, even though there are slurry flow measurements before the nozzles and power consumption measurements at the slurry feed pumps, it cannot be concluded if the spraying is functioning adequately or not at all the 9 nozzles per one reactor. Therefore, it would be beneficial if effect of spraying in the reactor could be monitored indirectly. As a result, indirect monitoring method of slurry injection was developed, which is described in this paper.

## 2 MODELLING OF SULPHUR REMOVAL

The proposed method to monitor functioning of spraying is based on energy balance. The basic idea is that by knowing the flows, contents and temperatures of injected flue gas, slurry and compressed air, the energy balance of the reactor can be calculated, if the reaction enthalpies of the chemical reactions taking place in reactors are small. With the energy balance, flue gas exit temperature of the rector, which is the main control variable in reactor control, can be estimated. The calculated temperature estimate indicates what should be the exit temperature if everything were in order. If the spraying is not functioning properly, the model output should differ from the measured values. E.g. if the drop sizes are too large, the temperature estimate should be smaller than the measured value indicating that the water in the slurry has not vaporized efficiently.

The challenge with this approach is, however, that there are a lot of variables that have an effect on flue gas and slurry compositions, but quite few of them are measured before the sulphur removal process in Salmisaari. Therefore, several variables have to be estimated based on measurement information and some additional information. In this approach, estimates that are only based on physical calculus are utilized to gain generality and traceability. This chapter describes model derivation.

## 2.1 Input flows to desulphurization reactors

### 2.1.1 Flue gas flow

The main input flow to SRP is flue gas from a power plant. In Salmisaari case, the power plant constitutes of two boilers; boiler K1 is a steam boiler with capacity of 160 $MW_e$ and 300 $MW_{th}$ and boiler K7 is a hot water boiler with capacity of 180 $MW_{th}$. Flue gases from these boilers are mixed before conduction to SRP. The main fuel of the boilers is pulverized coal, and heavy fuel oil is used as auxiliary fuel. Additionally, boiler K1 has started wood pellet co-combustion in turn of the year 2015, which is e.g. discussed in /5/. The boilers are under LCP and IED directives and EU Emission trading system, e.g. /7/, so the flue gas properties are measured thoroughly. However, the measurements are located after the SRP in the stacks (Figure 2), so there are only sensors prior to SRP that are used for process operation. Therefore, detailed flue gas properties must be estimated.

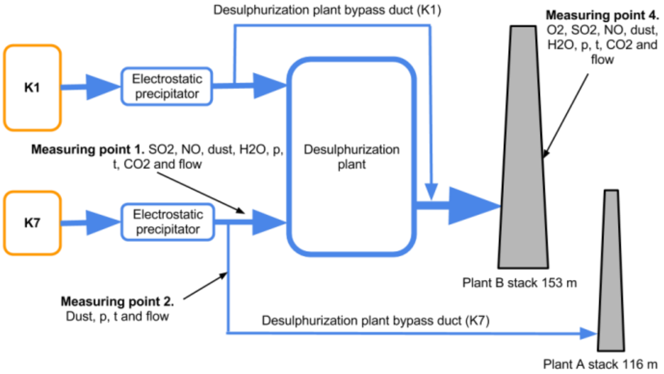

**Figure 2.** Locations of flue gas measurements /7/.

Flue gas composition can be estimated by boiler model presented in /6/ by applying the model to Salmisaari case with two boilers (K1 & K7) and three fuels (coal, heavy fuel oil, and wood pellet). There, fuel and prospective soot blowing steam flows and flue gas oxygen contents must be measured for both boilers separately. Additionally, fuel chemical compositions must be known, at least to some detail level. Air properties (temperature, pressure and humidity) can also be taken into account. With this measurement information, estimation of flue gas compositions and flows $\dot{m}_{FG,tot}$ of single boiler can be estimated. These flue gases can then be summed, and the total flue gas properties can be calculated.

There are two parallel reactors (R1 & R2) in SRP and the total flue gas (FG) flow is divided between them according to pressure conditions. Therefore, flue gas flow into each reactor can be estimated by

$$\dot{m}_{FG,r} = \frac{\sqrt{\Delta p_r}}{\sqrt{\Delta p_{R1}}+\sqrt{\Delta p_{R2}}} \cdot \dot{m}_{FG,tot}, \tag{1}$$

where $\dot{m}_{FG,tot}$ is the total flue gas mass flow estimate and $\Delta p_r$ the pressure difference in the duct of reactor $r$ = R1,R2. Heat power of the flue gas to reactor $r$ can be stated as

$$\dot{Q}_{FG} = \sum_i c_i \, \dot{m}_{FG,r,i} T_{FG}, \tag{2}$$

where $c$ stand for heat capacity and $T$ temperature. $i$ includes CO, $CO_2$, $H_2O$, $O_2$, $N_2$, $SO_2$ and HCl. Flue gas temperature before the reactors is typically 120–150 °C /4/, so heat capacity values at temperature 135 °C are used in the model.

### 2.1.2 Slurry flow

The slurry is mixed in dosing tank, and in steady state the slurry (SL) flow to reactor $r$ can be expressed as /4/

$$\dot{m}_{SL,r} = \dot{m}_{milk,r} + \dot{m}_{RCP,r} + \dot{m}_{CL,r} + \dot{m}_{water,r}, \tag{3}$$

where *milk* stand for lime milk (water and fresh $Ca(OH)_2$), *RCP* recycled solid product ($H_2O$, $Ca(OH)_2$, $CaSO_3$, $CaSO_4$, $CaCO_3$, $CaCl_2$, and ash; marked as $k$), and *CL* extra $CaCl_2$. The composition of slurry is such that density of slurry suspension $\rho_{susp}$ is at desired level, typically 1200 kg/m$^3$. The denser the slurry suspension, more Ca(OH)$_2$ is fed to reactors, which then promotes SO$_2$ removal. However, there is an upper density limit c. 1250 kg/m$^3$, and higher densities contribute to undesirable practical issues that e.g. deteriorate pumping of slurry. In practice, slurry composition is such that the amount of recycled solid end product in the slurry is maximized, and amount of fresh lime milk is controlled such that desired flue gas SO$_2$ concentration after the SRP is reached. Water is fed to control the slurry density to desired level at the dosing tank.

The density of slurry $\rho_{m,SL}$ in the dosing tank and volumetric slurry flows $\dot{V}_{m,SL,r}$ to reactors R1 and R2 are measured, which can be used to separate solid and liquid parts of the slurry feeds. In general, mass ratio of solid material over total suspension can be estimated by equation /4/

$$x_{solid} = \frac{\rho_{solid}(\rho_{m,susp} - \rho_{H2O})}{\rho_{m,susp}(\rho_{solid} - \rho_{H2O})}, \tag{4}$$

where $\rho_{m,susp}$ is measured suspension density and $\rho_{H2O}$ density of liquid water. In this case, density of solid substances involved are within range 2230–2330 kg/m$^3$, so it is estimated that average density of solid particles $\rho_{solid}$ is 2300 kg/m$^3$. When $\rho_{m,susp} = \rho_{m,SL}$, solid material and water flows to reactor *r* can be stated as

$$\dot{m}_{SL,solid,r} = x_{solid} \cdot \dot{m}_{SL,r} = x_{solid} \cdot \dot{V}_{m,SL,r} \cdot \rho_{solid} \tag{5}$$
$$\dot{m}_{SL,H2O,r} = (1 - x_{solid}) \cdot \dot{m}_{SL,r} = (1 - x_{solid}) \cdot \dot{V}_{m,SL,r} \cdot \rho_{H2O}. \tag{6}$$

In the calculus, the flow $\dot{m}_{SL,solid,r}$ is divided to components $k$ according to lime milk and recycled end product ratio. In the analysis, typical compositions of recycled end product and lime milk compositions were used.

Heat power of slurry flow can be formulated as

$$\dot{Q}_{SL,r} = \sum_k c_k \ \dot{m}_{SL,k,r} T_{SL} - \dot{m}_{SL,H2O,r} \cdot \Delta H_{vap}, \tag{7}$$

where the latter part considers liquid water in the slurry that vaporizes in reactors. There, the $\Delta H_{vap}$ stand for heat of evaporation of water.

### 2.1.3 Air flow

Slurry suspension is injected by compressed air through specific nozzles into the reactors. Heat power to reactor can be formulated as

$$\dot{Q}_{air,r} = c_{air} \dot{m}_{air,r} T_{air}. \tag{8}$$

The air flows to the reactors $\dot{m}_{air,r}$ are measured separately, but the air temperature measurement $T_{air}$ is not connected to automation system. Therefore, it is estimated with constant value, but due to modest air flows it has minor effect on power balance.

## 2.2 Output flow from the reactors

In the reactor, part of SO$_2$ and most of HCl are absorbed into the alkaline droplets to form calcium sulphite, sulphate and calcium chloride mainly with reactions

$$Ca(OH)_2 + SO_2 + H_2O \rightarrow CaSO_3 \cdot 0.5H_2O + H_2O \tag{R1}$$
$$CaCO_3 + SO_2 + H_2O \rightarrow CaSO_3 \cdot 0.5H_2O + CO_2 + H_2O \tag{R2}$$
$$Ca(OH)_2 + 2HCl \rightarrow CaCl_2 \cdot 2H_2O \tag{R3}$$

The total output power of the products escaping the reactor can be stated as

$$\dot{Q}_{out,r} = \sum_l c_l \, \dot{m}_l T_{exit,r} - \sum_l \dot{n}_l \, \Delta H^0_{f,l}, \tag{9}$$

where *l* covers CO, CO$_2$, H$_2$O, O$_2$, N$_2$, SO$_2$, HCl, H$_2$O, Ca(OH)$_2$, CaSO$_3$, CaSO$_4$, CaCO$_3$, CaCl$_2$, and ash. The output temperatures of the reactors are typically around 75 °C, so respective $c_l$ capacities at that temperature are used in the analysis. However, reaction enthalpies $\Delta H^0_{f,l,}$ of the reactions are minor compared to reactor input power, so for monitoring of flue gas exit temperature they can be omitted. When the chemical reactions are ignored, the chemical compositions of mass flows are the same as into the reactors, and the output flows of the reactors are flue gas flow to bag filters and solid mass flow to recycling

## 2.3 Power balance

By assuming adiabatic conditions in the reactor and combining equations 2, 7, 8 and 9, the total input and output (when ignoring the reaction enthalpies) powers to reactor $r$ can be set:

$$\dot{Q}_{in,r} = \dot{Q}_{FG,r} + \dot{Q}_{SL,r} + \dot{Q}_{air,r} = \sum_i c_i \dot{m}_{FG,r,i} T_{FG} + \sum_k c_k \dot{m}_k T_{SL} - \dot{m}_{SL,H2O} \cdot \Delta H_{vap} + c_{air} \dot{m}_{air,r} T_{air}. \tag{10}$$

$$\dot{Q}_{out,r} = \sum_l c_l \dot{m}_l T_{exit,r} \tag{11}$$

The energy balance can be stated as $\dot{Q}_{in,r} - \dot{Q}_{out,r} = 0$, and the solving the reactor output temperature $T_{exit,r}$ can be solved analytically e.g. with Matlab Symbolic Toolbox™.

# 3 MONITORING OF SO$_2$ REMOVAL − RESULTS AND DISCUSSION

Figure 3 presents a case example with 5 day period (120 hours). The simulation was conducted in Matlab Simulink™ environment with actual process data that was collected in typical plant operation during winter months. In the case, boiler K1 is run by coal almost at full load (Fig 3.1). Meanwhile, boiler K7 was started at the beginning of the period and flue gases of K7 are directed to SRP after 2 hours (Fig. 3.2). After full load operation, the boiler power of K7 was reduced before shut down at the end.

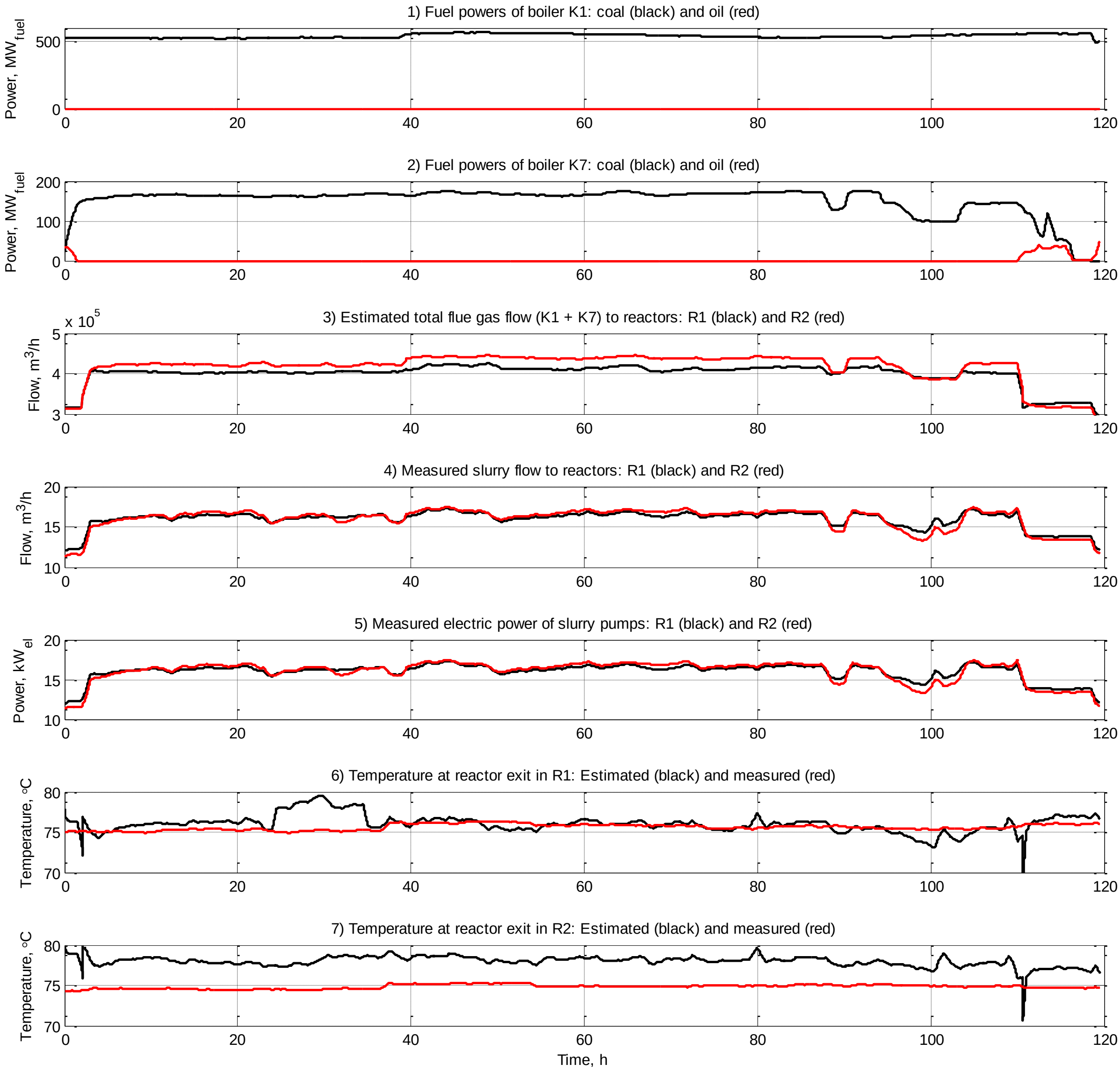

**Figure 3**. Simulation with actual process data. There is a simulated error in Fig. 3.6 at period 25–35 that illustrates situation that 5 % of the liquid in the slurry to R1 do not vaporize.

The Fig. 3.3 indicates that there is a pressure misbalance between the reactors and flue gas flow to R2 exceeds the level to R1 by almost 10 %. The measured slurry flows (Fig. 3.4) and slurry pump powers (Fig. 3.5) react similarly but by varying degree. As a result, temperature estimate and measurement at reactor R1 (Fig. 3.6) fit well with each other, beside an simulated error at period 25–35 (h) that illustrates situation that 5 % of the liquid in the slurry do not vaporize. In parallel, temperature estimate in R2 (Fig. 3.7) differ from measurement on average of 3 °C. The difference between the measurements can be due to measurement inaccuracies or process behaviour. As the slurry and flue gas properties are the same for both reactors and the same temperature drop in the reactor is achieved with roughly the same slurry flows, it is likely that other one of the pressure measurements that are used to separate the flue gas to R1 and R2 are biased, or alternatively there is an error in one of the slurry flow measurements. Still, the measurements and estimates fit fairly well in normal conditions, and the simulated error illustrates that the method is able to alert if there is something unexpected happening in the process.

In the analysis, the estimated trends can be compared with the measurements and between the two parallel and similar reactors to gain indication of changes at the reactors. Alternatively, the estimation can be made by different mode to calculate the required slurry flow that produces the measured flue gas exit temperature. Combination of estimates and measurements enable extended monitoring prospects by several new indexes that hopefully provide more insight to process behaviour that can be used in several applications.

## 4 CONCLUSIONS

This paper presented an indirect method to monitor spraying of calcium slurry in flue gas desulphurization process that was applied in Salmisaari power plant. The proposed method to monitor functioning of spraying is based on energy balance. With the energy balance, flue gas exit temperature of the rector, which is the main control variable in reactor control, can be estimated. The calculated temperature estimate indicates what should be the exit temperature if everything were in order. Due to lack of measurements before the SRP, several variables had to be estimated based on measurement information and some additional information. Still, the results verity that the method is able to predict the reactor exit temperature by error of typically less few degrees Celsius regardless of process state.

In the presented approach, estimates that are only based on physical calculus were utilized to gain generality and traceability. The benefit of this approach is that information from several sensors is connected to form the estimates, but the drawback is that inaccuracies in measurements affect the estimated result. Therefore, the tools also monitor the measurement systems and possible abnormal situations can also be explained by non-optimal measurement performance. Still, the generality of the method is beneficial, as e.g. coal types and power levels with two boilers vary constantly. Moreover, the process operations and therefore estimation challenges are further increased after the application of wood pellet and coal co-combustion taking place in Salmisaari and Hanasaari power plants in 2015. As the monitoring method uses only measurements and general process information, the method should work without significant modifications also with the new operation environment.

## 5 ACKNOWLEDGEMENTS

The work presented in this paper was conducted in Measurement, Monitoring and Environmental Assessment (MMEA) program managed by CLEEN Ltd and primarily financed by Tekes. Project financers and partners, especially Helen and IndMeas, are gratefully acknowledged.

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