Decentralized storage in combined heat distribution circuits: how to control?

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Abstract. In apartment buildings, collective heating networks have great energetic advantages. One form gaining more attention for the last decades is a combined heat distribution circuit (CHDC), in Belgium called “combilus”. It is a two-pipe system for the distribution of both space heating (SH) and domestic hot water (DHW). The supply temperature is set to the highest needed temperature, which is around 65°C for DHW (considering a temperature difference for enabling heat transfer). However, if decentralised DHW storage tanks in combination with low-temperature emitters for SH are used, the supply temperature could be optimised. In this research, two innovative control strategies were studied for such a CHDC to lower the distribution temperature (to the required temperature for SH) as much as possible by grouping the charging periods of those storage tanks. One control strategy is time-based, with pre-defined charging schemes, while the other is based on two sensors in the storage tanks. In order to test the control strategies, a simulation environment was set up in Matlab that represents the thermal dynamic behaviour of CHDC. However, to fully focus on the evaluation of the control strategies, an idealised central boiler room was assumed, which immediately delivers the desired temperatures. Besides the evaluation of the control strategies, the design of the storage tanks is also optimised by performing sensitivity analyses on the volume, hysteresis and charging flow rates. The results show that larger storage tanks provide better DHW comfort combined with less PE use (for the proposed controls) and that the charging flow rate can significantly reduce the central peak power, while DHW comfort and PE use remain the same. With the time-based control, the charging cycles and volume have a high impact on the performance and comfort. The two-sensor control is always able to reduce PE use and deliver the same or even better comfort than the reference control.

Keywords. Domestic hot water, decentralized storage tanks, combined heat distribution, control strategy, collective heating.
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1. Introduction

1.1 Combined heat distribution circuits

Combined heat distribution circuits (CHDC) are collective heating systems in apartment buildings with only one supply pipe and one return pipe (2-pipe system). These pipes distribute heat for both space heating (SH) and domestic hot water (DHW) to the dwellings. By using only one supply pipe, the distribution losses are reduced compared to a 4-pipe system. In Dutch, this system is called “combilus”.

Currently, the supply temperature in a CHDC is set at 65°C, the hottest demand, to guarantee the supply of DHW to the end-users at all times without major waiting times. However, this leads to two main disadvantages. Firstly, distribution losses are mostly higher than strictly needed. Secondly, the overall energy efficiency decreases because of high return temperatures [1] when the overflow valve is opened.

Another incentive to regulate the supply temperature in a more adequate way is that the share of SH demand decreases in importance due to increased insulation [2-3]. Low-temperature emitters, such as underfloor heating and convectors, are becoming more common in the new-built environment. This means that low temperatures can be distributed in the CHDC, if an energy and comfort-friendly solution is provided for the DHW supply. The most promising solutions are decentralised storage tanks [1, 6] and booster heat pumps [6-7] for DHW storage and production at local level, respectively. This research focuses on using decentralised storage tanks for DHW in CHDC.

1.2 Decentralised storage tanks

Decentralised storage tanks are small thermal storage tanks in every dwelling to store DHW. With an internal coil heat exchanger (CHE), the heat from the CHDC is transferred to the water in the tank. This heated water is the consumable DHW for end-users.

Today, decentralised storage tanks are already used in CHDC or district heating networks. However, they are not yet used in combination with an optimised supply temperature control strategy (still set at 65°C), but rather to reduce waiting times for DHW use and make overflow valves unnecessary [8].

Combining low-temperature emitters with decentralised DHW storage tanks provides opportunities for demand-based control strategies in CHDC. During most of the day, the supply temperature could be lowered for SH and when the DHW tanks need to be charged, a temperature of 65°C can be distributed. This has two main advantages: I) reduced distribution losses and II) increased total production efficiencies as low temperatures can be generated by renewable energy sources, such as heat pumps (HP) [7].

1.3 State-of-the-art

Currently, many studies on temperature control optimisations and on decentral storage tanks exists in the scientific literature. Gustafsson et al. [9] investigated the possibilities of supply temperature control in district heating networks to increase the temperature difference between supply and return. They confirm that supply temperature control is usable in collective systems, while guaranteeing comfort. However, the temperature difference was hardly improved. Here, the DHW demand was neglected and it was based on in-situ measurements. In study [1], the performance of six configurations of heat interface units (HIU) in CHDCs and different control strategies were compared, based on return temperature, heat losses and DHW use temperature. However, the control strategies were applied to the overflow valve for recirculation of the CHDC and radiators were considered for SH. Another study [10] presented a general overview of possible concept for DHW production in low-temperature heating systems. A novel control strategy was also introduced, but the focus was again the recirculation loop, and no decentralised storage tanks were considered. In addition, [12] developed an active control strategy with data-based techniques for a district heating network with a CHP. They compared different set-ups, amongst others central storage tanks or decentral storage tanks. The decentral storage tanks yielded the highest possible savings.

Besides proposing new control strategies, the effect of different design choices in the storage tank should be taken into account. As stated in [11, 13], the design has a major impact on the energy performances. Van Minnebruggen et al. [14] validated a dimensioning tool for thermal storage and required heating power in collective heating networks, but the focus lies on the central boiler room, rather than the decentralised storage tanks. In [7], the use of booster heat pumps in combination with decentral storage was assessed for CHDCs. In this study, the focus was on the efficiency of the system by using only heat pumps and on the influence of sizing. The decentral DHW production and storage lead to great energy savings in comparison to CHDCs without decentral storage.

The state-of-the-art lacks optimised control strategies for CHDCs with decentral storage and an evaluation of design choices on the performance of control strategies in CHDCs.

1.4 Outline

The following section explains the used methods and the main principles/assumptions of the simulation environment. Section III is devoted to the description of the proposed control strategies for the supply temperature and the different sensitivity analyses for the storage tanks' design. Afterwards, in section IV, the results for the proposed control strategies are given, with a discussion. Finally, this paper concludes with the main findings of this study.

2. Research Method

2.1 Simulation-based

A dynamic simulation environment is built in Matlab to test control strategies and study the influence of different design choices. By doing so, the boundary conditions and heat demand for DHW and SH could be kept the same for an objective evaluation. The models are based on the doctoral dissertation of Van Riet [15] and are also used in [7,16-18].

This environment represents the dynamic thermal behaviour of the CHDC and its components. The transient behaviour is taken into account by differential equations according to the general equation described in [16]. Only the storage tank model is a partial differential equation both in temperature and along the height (temperature layers). The simulation time step is 10 seconds to have detailed simulations of the DHW use. It is assumed that the CHDC is perfectly balanced, thus the desired mass flow rates are always available. The time-delay in the pipes is taken into account by applying the plug-flow principle [18].

Due to the simulation time step of 10 seconds and the detailed simulation models, the simulation period has been fixed to only one month (January) to reduce the computational time and power.

2.2 Case study

The apartment building of this research consists of 20 identical dwellings, located in Uccle, Belgium. All dwellings have a storage tank for DHW and SH is based on underfloor heating with design temperatures 35°C/30°C. No hydraulic separations (e.g. heat exchangers) are needed as the apartment building is small (no high pressures in CHDC) and the storage tanks separate the DHW from the technical water in the CHDC. All storage tanks have a priority switch for DHW, thus if a tank is being charged, the SH in that dwelling is shut off. The indoor temperature set point is 21°C during the day and 19°C at night. The heat load by design conditions (21°C indoor and -8°C outdoor) is 1340 W. Each dwelling has one shower and two or three other tapping points. They are all occupied by different families of one to three residents. The internal heat gains, occupancy profiles and DHW demand profiles are based on a stochastic "profile generator" developed in the Instal2020 project [4-5]. An example of the DHW profile for all dwellings for two days is given in the appendix to this paper (Fig. A).

The central boiler room consists of a geothermal heat pump (GHP), connected to a storage tank, and a boiler in series. The boiler room is considered as ideal, thus it can instantaneously deliver the demanded supply temperature and mass flow rate by the control strategy. This approach is chosen, as it allows us to focus on evaluating the control strategy, without the effects of transient behaviour in production units or incorrectly set PID controllers. An overview of the case study is shown in Fig. 1.

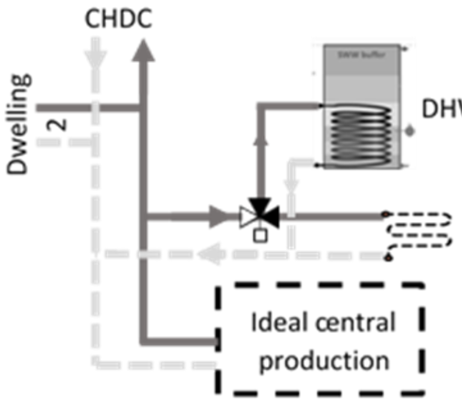

Fig. 1 – Overview of the case study. The ideal central production consists of a geothermal heat pump and a boiler. Every dwelling in the CHDC has a storage tank for DHW and underfloor heating.

2.3 Model of DHW storage tanks

The stratified thermal storage tank has an internal coil heat exchanger (CHE) in the lower half of the tank and two ports at 0% and 100% of the height. The model is based on Type 60 of TRNSYS [19] and described in [15]. The tank is divided in different homogeneous water layers. Heat transfer to the adjacent layers due to conduction and advection is considered, as well as losses to the surroundings and heat gains from the internal heater. A temperature inverse algorithm is added to take account of the buoyancy effect. At the in- and outlet, no account is taken of buoyancy, but rather of mixing. It is possible to add an electrical resistance in one of the layers. The dimensioning (UA-value [W/K]) of the internal CHE is based on lab measurements of a 90 l storage tank and technical information from *Collindi* [20]. When larger volumes are simulated, a larger coil with increased nominal power is foreseen and UA is modified as such. For different charging flow rates, a constant UA value is assumed, which means that the internal CHE is smaller and longer.

2.4 Key Performance Indicators

To compare the control strategies and analyses, four key performance indicators (KPI's) are used:

First is the total primary energy (PE) use of the system (PE_{use}[kWh]). The PE use of the ideal central boiler room is calculated and, if any, summed up with the PE use of the electric resistances in the storage tanks. The GHP's set point is 45°C. Its Coefficient of Performance (COP) at 45°C/37°C is assumed to be 4.8 and adjusted to the return temperature (T_{ret} [°C]) according to the EPB calculation method [21], shown in equation (1)). If the supply temperature is above 45°C, the boiler will provide the extra heat with an efficiency of 100%. The conversion factor for electricity to PE is 2.5 in Belgium. This KPI is important since this research aims at reducing the energy use.

$$COP = 4.8 \cdot (1 + 0.01 \cdot ((45 - T_{ret}) - 8)) \qquad (1)$$

Next is the Primary Energy Ratio (PER). This is the total efficiency in terms of PE, i.e. the ratio of PE_{useful} to PE_{use}. PE_{useful} [kWh] is the useful energy for SH and DHW. A higher PER indicates a higher heat pump's share in energy delivery and lower heat losses.

The relative duration of DHW temperature lack [%] is introduced as a KPI. This is the proportion of the total tapping times of all storage tanks that the DHW temperature is lower than 40°C. This KPI refers to the situation where the storage tanks are getting empty at the end of a tapping period. In contrast to a temperature lack that occurs at the beginning of a tapping, which is related to pipe lengths, this discomfort is due to sizing or late recharging of tanks (control strategies). The smaller this percentage, the less time the end-users experience discomfort on average. The distribution pipes from the storage tanks to the tapping points are not considered.

Finally, the central peak power (CPP) [kW] is used as KPI. This is the highest power delivered by the central boiler room over a time period of 10 minutes. This larger timeframe is chosen to have more realistic values, since an ideal production without inertia – and thus providing unrealistic temperature increases - is assumed. The lower this peak power, the smaller the boiler and GHP can be designed, which reduces investment costs and increases the overall energy performance.

3. Experiments

3.1 Control strategies and sensitivity analyses

The control strategies are considered for a CHDC that can operate at both low (38°C) and higher (65°C) temperatures. A straight forward control strategy is to raise the distribution temperature each time one of the storage tanks demands heat. Although the temperature can be lowered outside these charging cycles, simulations show that low distribution temperatures rarely occur for such a basic control strategy. Hence, the proposed control strategies are meant to increase the simultaneity of the charging storage tanks and thus minimise the time period with high distribution temperatures. As a result, primary energy use is expect to decrease as described in 1.2.

The impact of two optimized control strategies on energy use and DHW comfort is assessed and compared to the reference control strategy, where the distribution temperature is high (65°C) both for SH and DHW demand. The design parameters are set according to a case study measured in Malle [22]: the reference volume is 90 l (for one shower per dwelling). The tanks have one sensor at 2/3th of the height and a set point of 58°C with an ON/OFF control hysteresis of +- 3°C. The charging flow rate of the storage tanks is initially set at 600 kg/h.

The first optimized strategy is a time-based control. The distribution temperature is raised at some pre-defined time slots and all tanks, with demand according to one sensor at the bottom of the tank, are thermally loaded. It is important to define the size of the storage tank and the charging time slots to ensure DHW comfort in between. Therefore sensitivity analyses are performed for these parameters.

For the second strategy, two sensors are used in each tank. If there is a demand according to the upper sensor of one tank, distribution temperature is raised and all tanks with demand according to their lower sensor are loaded simultaneously. This control strategy can be optimized by improving the hysteresis and the size of the storage tanks, which is also subject of this research.

Due to the increased simultaneity, the CPP is expected to increase with the proposed control strategies. Therefore the impact of lowering the charging mass flow is assessed for both strategies.

4. Results and discussion

4.1 Time based control (TB-control)

Fig. 2 shows PE and DHW discomfort for January when a time based control (TB-control) is used, compared to the reference as described in 3. In the following figures, the shape refers to the volume of the storage tank (150, 200 or 300 litre), the colour to the charging flow rate (from 100 kg/h to 600 kg/h). For each combination of volume and flow rate, 5 different time slots are considered, with a total charging time of 1, 2, 3, 4 or 6 hours. This total charging time is divided equally between a morning and evening charging cycle, because this proved to give better comfort results almost without affecting the energy consumption. So, the rightmost datapoints, with the largest PE, are for a charging time of 2*3h. As the charging time shortens, energy use decreases and the points move to the left on the graph. They are indicated with smaller data points. Finally, it should be mentioned that the data points for the 3 volumes, with 100 kg/h and a 2*0.5h charging cycle, have discomfort percentages up to 65% and fall outside the limits of the graph.

Fig. 2 makes clear that, compared to the reference case, TB-control leads to lower energy use, while DHW discomfort highly depends on the storage volume and the duration of charging cycles. For 300l storage, a charging time of 2*1 h (>=300 kg/h) or 2*1.5 h (100 kg/h) results in nearly no DHW discomfort. Extending the charging time is not recommended, as this only leads to higher energy use. For smaller storage volumes, and the considered time slots, there is always some discomfort. However, it is possible to maintain at least the comfort level of the reference with a charging time between 2*1 h and 2*2 h (depending on volume and flow rate). For smaller volumes, extending charging times or increasing flow rates improves DHW comfort, but despite higher energy use.

Given the limited place in apartments, increasing the storage temperature (+10°C) might be preferable instead of larger volumes. Another way to provide a better DHW comfort for smaller storage volumes, is adding an electrical resistance (ER) in the storage tank. These datasets are added in Fig. 3, outlined in red and in green, resp. Only the cases with a flow rate of 100 kg/h are included here (for readability). Fig. 3 shows that taking these measures do indeed improve DHW comfort, but at the expense of increased PE use. The increase in PE is limited when increasing the storage temperature, but when local ERs are added, the total PE exceeds even the reference control. It should be mentioned that this conclusion highly depends on the assumed PE conversion factor for electricity (for Belgium; 2.5). If PV panels are present, the cases with local ERs will be more advantageous.

The TB- strategy is intended to combine the charging cycles of the different storage tanks and therefore result in higher CPP (see Fig. 4) compared to the reference. However, also a significant reduction of CPP is possible when the charging flow rate of the storage tanks is reduced. This also results in PE saving (Fig. 2 and Fig. 4). It is concluded that, based on PE, DHW comfort and CPP, the best option is to work with reduced flow rates (100 kg/h) and a volume of 300 l with a charging time of 2*1.5 h. However, good results can be obtained with smaller volumes, but charging time should be adjusted to the other parameter settings and comfort requirements.

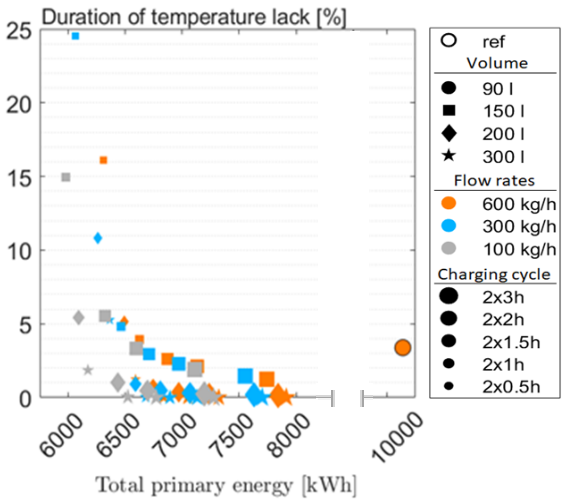

Fig. 2 – TB-control: PE and DHW discomfort for different storage volumes, flow rates and charging cycle.

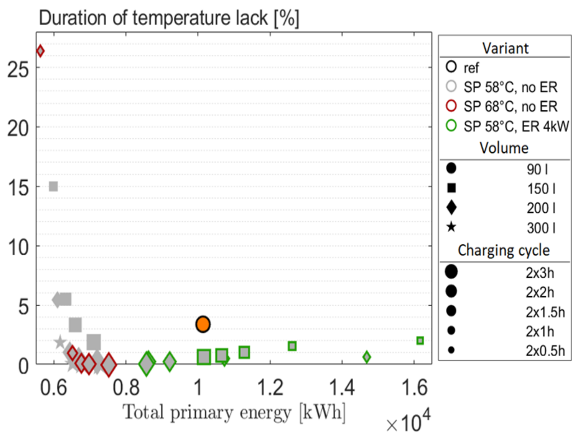

Fig. 3 – TB-control: PE and DHW discomfort, variants on storage temperature and electrical resistance.

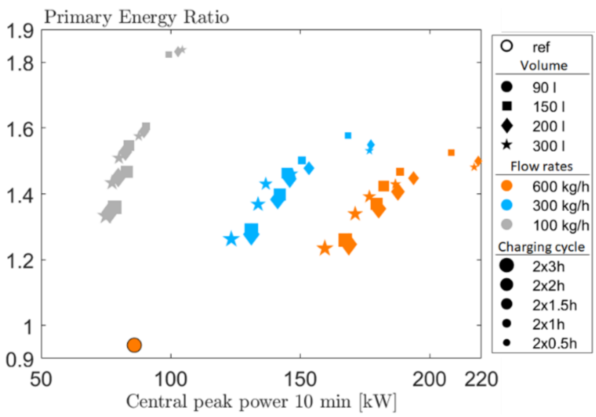

Fig. 4 – TB-control: central peak power and PER for different storage volumes, flow rates and charging cycle.

4.2 Two-sensor control (2SC)

The second proposed control strategy, based on two sensors, is called "two-sensor control" (2SC). The set point of the sensors in the storage tanks is always 58°C. In following figures, the shape refers to the volume (90, 150 or 200 litre), the colour to the charging flow rate (from 100 kg/h up to 600 kg/h) and the darkness of the (same) colour refers to the hysteresis (light colour variant is +-3°C, the darkest variant of a colour is +-5°C). These variations on the reference control strategy (always supply temperature of 65°C and with a hysteresis of +-3°C) are also given, slightly outlined in black.

Fig. 5 shows for all variants the PE use vs. the discomfort of DHW in % of tapping time (see section 2.4 for exact definitions). A few conclusions are drawn from this chart. Firstly, the influence of the volume on the discomfort for DHW is clearly visible. For both the reference and the 2SC, a larger storage tank consequently improves the comfort level for DHW. While the reference control requires approximately the same PE (same charging flow rate considered, i.e. colour), and the 2SC reduces PE use by up to 25% (when the charging rate is 600 kg/h for 200 litre tanks) compared to its reference. In fact, with the 2SC, PE savings are achieved for each variant, while DHW comfort is roughly the same or even better than with the reference control. The PE use is similar for different tank sizes in reference control, because of similar storage losses (all between 3 and 4% of the total energy demand). The PE savings with the 2SC is due to the simultaneous charging, as the supply temperature is low during longer periods for larger tanks.

Secondly, for the 2SC, the charging rate has nearly no influence on the comfort and PE use for the larger storage tanks. For the 90 l tanks, the discomfort increases when using a smaller charging rate and the PE use is slightly smaller. In contrast, the charging rate has a high influence on the PE use of the reference control. The effect on DHW comfort is also larger for smaller tanks in reference control. These different results are due to simultaneous charging of tanks. With the 2SC, many tanks are simultaneously

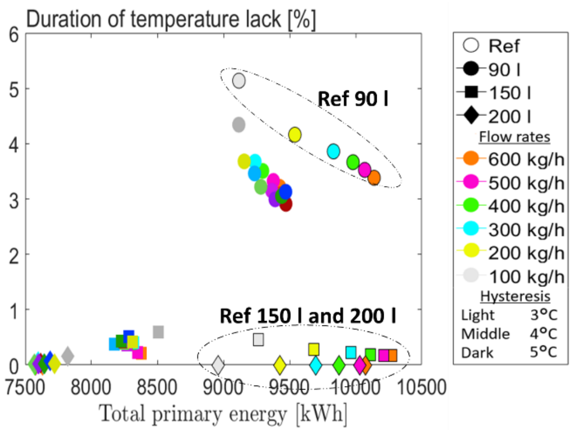

Fig. 5 – Two-sensor control: Pareto analysis on the DHW discomfort [%] and total PE use [kWh] for different charging rates, volumes and hysteresis.

charged. When the charging rate decreases, the return temperature during charging is lower, which increases the production efficiency. However, the smaller charging rate leads to a high distribution temperature for longer periods. The increased distribution losses and efficiency gains cancel each other out for 2SC, resulting in similar PE uses. In reference control, the discomfort increases, since small charging rates are not sufficient, certainly not for small tanks. The PE use decreases due to higher production efficiencies (lower return temperatures).

Finally, the hysteresis (only shown for two-sensor control) does not seem to have a pronounced effect on the larger volumes, but on the 90 litre tanks it can be used to optimise the DHW comfort.

Now the different influences on comfort and PE use are clear, the influences on the required central peak power (CPP) over a period of 10 minutes in relation to the PER is examined in Fig. 6.

For the 2SC, the required CPP increases with higher charging flow rates and larger storage tank volumes. The larger the tank, the larger the relative reduction for smaller charging rates is. The standard deviation of the PER for each volume separately is low from 600 to 300 kg/h. For larger tanks with a charging rate of 200 and 100 kg/h, the PER decreases. The effects on the PER are related to the PE use from previous discussion (Fig. 5). In reference control, the CPP also decreases with a decreased charging rate, but to a lesser extent than in 2SC. Due to the non-simultaneity charging moments, the effect of the flow rate is smaller. On the other hand, the PER increases in a greater extent, which is also consistent with the findings on PE use (Fig. 5).

For the 2SC it is possible to indicate an optimal variant for every volume of the storage tank. For 90l, a charging flow rate of 400 kg/h with hysteresis of +-4°C, is preferred. The comfort is better than the reference control, with the least PE use. For 150 l and 200 l, smaller flow rates will slightly decrease DHW comfort (but always less than 0.6% of tapping time), while the CPP decreases significantly (up to 54%). Thus, a charging rate of 100 kg/h is preferred. For the reference control and the best variant of the 2SC,

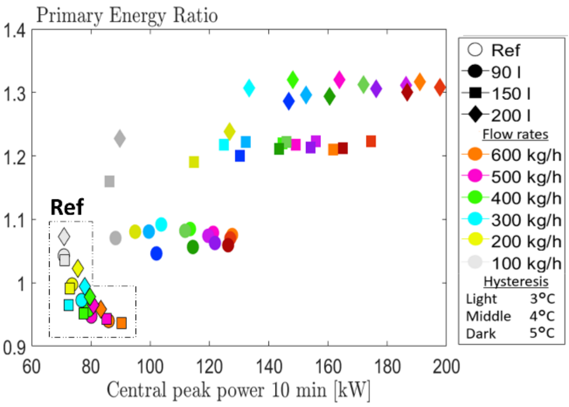

Fig. 6 – Two-sensor control: Pareto analysis on PER and central peak power.

Fig.7 presents the influence of the number of dwellings on both PE use and DHW discomfort.

As the number of dwellings decreases, the 2SC makes it more difficult to maintain a good DHW comfort. The reason is a decreased probability of a top sensor in a particular tank that gives a signal. Thus, the CHDC will supply more often low temperatures and tanks will be recharged less frequently, resulting in smaller effective storage volumes. On the other hand, the relative PE savings for an apartment building with fewer dwellings is larger, because of these lower supply temperatures. As expected, the central peak power decreased with the number of dwellings, while the PER increases slightly.

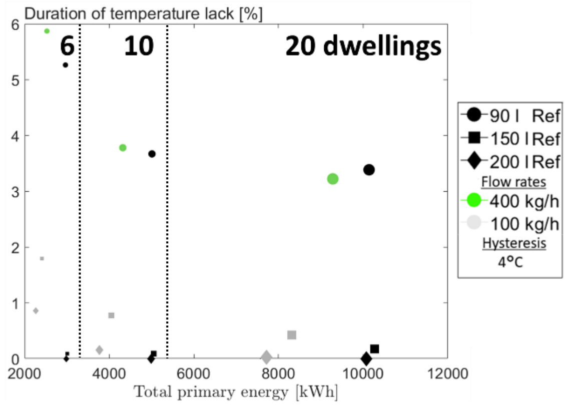

Fig. 7 – Influence of number of apartments for best variants of 2SC and on the reference control.

4.3 Comparison of control strategies

In 4.1 and 4.2 is shown that optimized control strategies allow to decrease PE use and at the same time, achieve a better DHW comfort. Those refined control strategies require sufficient attention to the settings of the significant parameters. Fig. 8 summarises the results. While the TB-control saves more energy with smaller charging cycles, it impacts the DHW comfort, and larger volumes are required (a compromise must be found between these KPI's). For the 2SC it is possible to specify an optimal variant for each volume, and saving more energy with increasing volume.

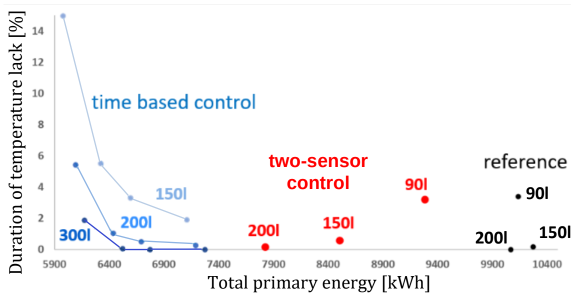

Fig. 8 – DHW discomfort and PE use for reference control (600 kg/h) and proposed control strategies (100 kg/h).

As mentioned before, all analyses were performed considering an ideal central boiler room. It is expected that PE savings would be less for a realistic boiler room, because the losses in the boiler room, and the delay of valves etc. are not considered. In addition, the efficiency of the boiler is set at 98%, while the COP of the GHP is calculated as done in EPB [21]. Besides these technical simplifications, the distribution temperature would not immediately be increased from 38°C to 65°C in reality, and thus this would also affect the DHW comfort. However, the goal was to compare different control strategies and these assumptions were made for both the reference control as the proposed control strategies.

4. Conclusions

Currently, the supply temperature control of CHDCs is set at 65°C to supply DHW at all times. However, with DHW storage tanks and low-temperature emitters in the dwellings, there are possibilities to optimise this supply temperature control. The goal is to reduce the supply temperature to the SH design temperatures for most of time by grouping charging times of all storage tanks as much as possible.

In this respect, this research proposes two control strategies. The first is a time-based control strategy, where one or two charging moments are pre-defined. The second one is based on two sensors. If the upper sensor in one of the tanks measures a too low temperature, the central supply temperature is set to 65°C. All storage tanks that are too cold at the bottom sensor will be charged. Afterwards, the central supply temperature is set back to 38°C for SH (design temperature of underfloor heating is 35°C/30°C). Both controls have a priority rule for DHW, so no SH is enabled in a given dwelling when its storage tank is being charged. Besides new control strategies, sensitivity analyses were performed on storage volumes, sensor's hysteresis and charging flow rates.

In order to test the two control strategies and to compare it to the reference control, a simulation environment is built in Matlab. The models are based on previous research [7, 15-18] with differential equations to simulate the dynamic thermal behaviour of the system. The central boiler room is considered as ideal, so it delivers directly the desired temperature to the CHDC. The DHW profiles, internal heat gains, occupancy patterns are based on a profile generator from Instal2020 [4-5]. The simulation time step is 10 seconds to simulate DHW demand in detail.

The proposed control strategies offer opportunities to lower PE use in CHDCs. At the same time, choosing the right settings to preserve DHW comfort and to limit peak demand, could pose some challenges. In summary, the main conclusions of performed analyses are: I) increase the volume of storage tanks, as this will increase the DHW comfort level, while the total PE use is similar for the reference control and decreases even further for the proposed controls. II) The time-based control strategy is quite simple to introduce and has the potential to perform better than the reference control. However, an under- or overestimation of charging time and/or flow rate will have significant (negative) effect on resp. DHW comfort or PE. Besides, larger volumes (than 90 l) are required to provide the DHW comfort. III) The two-sensor control reduces the PE use (up to 25%), while delivering similar or improved DHW comfort (less than 0.6% discomfort). However, the control is way more complex to implement. IV) If one of the proposed control strategies is applied, the charging flow rate should be reduced to lower the central peak power, and thus reducing the investment cost of the central boiler room and the sizing of the piping. By reducing the flow rate from 600 kg/h to 100 kg/h, half the peak power is required, while maintaining the level of comfort with a similar PE use.

In future work, the position of sensors for the 2SC could be analysed. It is expected that these sensors will influence the level of comfort, but also the optimisation potential. Concerning the TB-control, different DHW profiles should be investigated. It is expected that a more flattened profile would complicate this strategy. The proposed control strategies should also be tested in a simulation environment with a realistic central boiler room to research the influence of thermal inertias and the influence of PID controllers. Finally, using artificial intelligence to optimise control strategies is an interesting follow-up research.

5. Acknowledgements

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6. Appendix

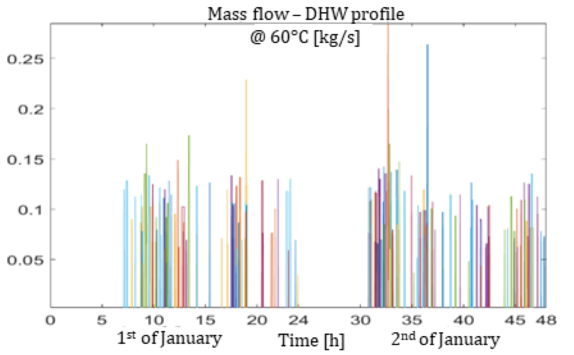

Fig. A – Example of DHW profile for the 1st and 2nd of January. The colors represent the different dwellings.

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Data Statement

The datasets generated and/or analysed during the current study are not publicly available because it is part of ongoing research, but are/will be available on request by mail to the corresponding author.