1 INTRODUCTION

Growing awareness of the ongoing climate change has led to increasing research activity in the field of CO_2 mitigating technologies. According to the current status of knowledge the emissions of carbon dioxide and other greenhouse gases (GHG) need to be reduced significantly in order to limit global warming. There is a broad political consensus that the global temperature rise should be limited to two degrees Celsius, compared with preindustrial temperatures. However, up to today there is no binding successor to the Kyoto Protocol under which 37 industrialised countries committed themselves to a reduction of GHG emissions by 5.2 % from the 1990 level until the end of 2012.

If the global temperature increase is to be limited to below 2 °C – a goal that has recently been affirmed by the United Nations Climate Change Conference in Copenhagen in December 2009 – global GHG emissions need to be reduced significantly [1]. As 41 % of global GHG emissions result from the combustion of fossil fuels for electricity generation, power plants play a critical role in the struggle against global warming [2].

Three fundamental paths exist to achieve a reduction in the emissions of GHG emanating from electricity generation:

1. change to less carbon-intensive energy sources such as natural gas, nuclear power, and in particular renewables;
2. increase in energy efficiency both in electricity production and consumption;
3. capture and storage of the produced carbon dioxide from fossil-fired power plants.

It is predicted that renewables and nuclear energy will only provide part of the world's energy needs in the next decades and that fossil fuels will remain a key energy source. Of all fossil fuels, coal has the largest resources and shows a wide global distribution of reserves. The continuing use of coal ensures a diversification of the energy supply and thus safeguards security of supply, especially in countries lacking their own natural gas and oil resources. 7,756 TWh_{el} or 41 % of global electricity generation in 2006 originated from coal where the share is predicted to increase to 44 % by 2030. In the EU

1,021 TWh_{el} (31 %) originated from coal-fired power plants in 2006. In China electricity generation from coal has more than doubled between 2000 and 2006 to a total of 2,328 TWh_{el} (80 %) [2].

GHG emissions from coal-fired power plants can be reduced by increasing the energy conversion efficiency of these plants or by separating the emanating CO_2, commonly referred to as carbon (dioxide) capture and storage (CCS).

CO_2 emission reduction by efficiency increase offers the benefit of reduced fuel consumption while keeping the net power generation constant. Yet, the resulting reduction in emissions is limited. Current efficiencies of coal-fired power plants have reached a plateau at which further efficiency increases demand a major effort in material development in case of conventional steam power plants or the commercial introduction and optimisation of the integrated gasification combined cycle (IGCC) [3].

CCS would permit the continuing use of coal and other fossil fuels while significantly reducing GHG emissions. CCS has been discussed since the 1980s, but the lack of economic incentives as well as political and legal uncertainties have only allowed for the realisation of a few pilot plants worldwide and recently for the planning of some demonstration plants [4]. Besides the technical and economic aspects, safety and regulatory issues with respect to transport and storage of CO_2 remain unclear. Until today the concept of CCS for coal-fired power plants has not been realised on a large scale.

To inject CO_2 into adequate storage sites such as saline aquifers or depleted oil and gas fields, it needs to be separated from other gas components. Three technology routes exist to perform this separation. Figure 1.1 shows a schematic of each of the three technology routes that are commonly referred to as post-combustion, pre-combustion, and oxyfuel[1].

Even though pre-combustion and oxyfuel processes are considered to suffer a less significant decrease in efficiency when applied to power plants, post-combustion CO_2 capture (PCC) is technologically more mature. Commercial experience from gas treating applications is available at large scale in industry and existing power plants can be retrofitted more easily [5, 6].

[1] For a detailed explanation of all three CCS technology routes refer to [3].

There is a large number of separation technologies for PCC in coal-fired steam power plants, but it is agreed that the implementation of an absorption-desorption-process using a chemical solvent is the most developed and adequate process for deployment in the near- to middle-term [7].

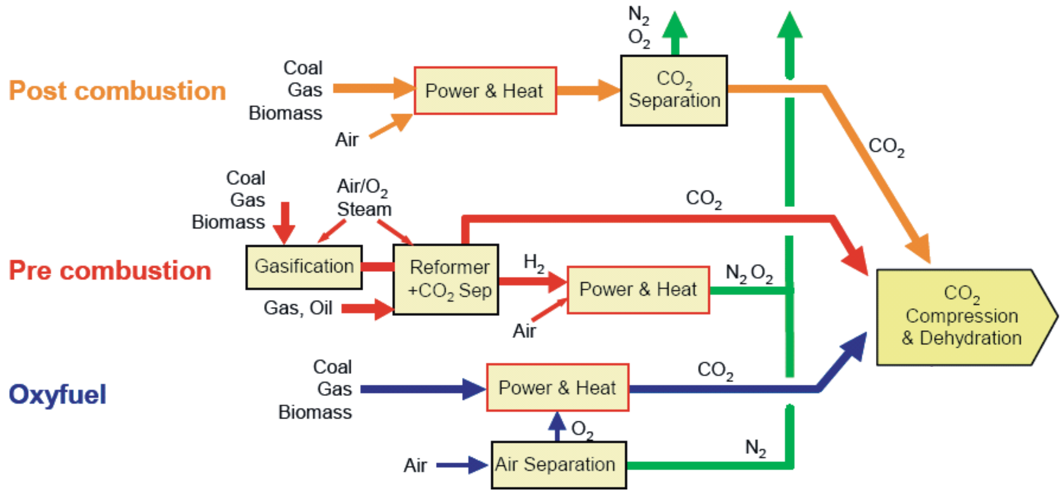

Figure 1.1: Technology routes for CO_2 capture [5]

1.1 Background

The application of a PCC process in a coal-fired steam power plant is associated to a significant loss in power output and a related net efficiency penalty of 8–12 %-pts. Besides considerable capital expenditures for engineering, procurement, and construction of a PCC process, the loss in power output leads to significant additional costs for the operation of a coal-fired steam power plant with CO_2 capture. Thus, an optimisation of the overall process and a reduction of the efficiency penalty is required for the commercial deployment of this technology and the application at industrial scale. An integrated model of the overall process would allow evaluating the impact of the choice of chemical solvent, process configuration, and operational parameters on the performance, thus on the net output, of the power plant.

The performance of state-of-the-art steam power plants and the impact of optimisation measures can be evaluated with a high degree of accuracy by using modern simulation tools that have been developed for this particular purpose. PCC by chemical absorption can be modelled with the help of equally powerful simulation tools developed for chemical and process engineering purposes. Accurate models of steam power plants as well as of CO_2

capture processes are, however, of high complexity. Consequently, simulations are computationally intensive and a direct link between the two models is intricate. Such connection is hence either infeasible or does not allow to consider all key process parameters in the optimisation of the overall process with reasonable effort and within a reasonable timeframe.

1.2 Aim and scope

Due to the increasing interest in CO_2 capture technologies in the power industry, research concerning the energetic evaluation of chemical solvents for CO_2 capture from flue gas has grown considerably in the past years. The majority of research is found on the fundamental physicochemical properties of novel chemical solvents such as CO_2 solubility, viscosity, density, or heat capacity. Industrial and academic research activities make use of the results of the fundamental research to develop models for the representation of absorber [8, 9, 10, 11, 12] and desorber (stripper) [13, 14, 15] columns.

In a smaller number of scientific publications the complete CO_2 capture process is evaluated with a focus on the minimisation of its energy requirement [16, 17, 18, 19, 20, 21]. NOTZ *et al*. developed a short-cut method for the energetic assessment of solvents for PCC within the CO_2 capture island [22]. The method uses a modified Kremser equation which requires a linearised relation for the representation of CO_2 solubility as a function of temperature and CO_2 concentration in the liquid. Additionally, the simplified assumption of a constant gas flow rate over the whole column is required for the application of the Kremser equation. In some studies the interaction of the capture process with the power plant is estimated by employing simplified correlations which relate the required energy to the corresponding loss in power output of the power plant [23, 24].

There exists a variety of studies that focus on the optimal integration of PCC processes in power plants. The integration in gas-fired combined cycles is, for example, discussed in [25, 26, 27]. Due to the significant differences in the boundary conditions of such power plants (e.g., different steam power process configuration, lower CO_2 partial pressure in flue gas) the transfer of the results of these studies onto coal-fired steam power plants is limited. The integration of CO_2 absorption processes in coal-fired steam power plants is also covered in literature [28, 29, 30, 31]. Many studies, however, use simulation tools which are not specifically designed to represent the complex interrelations in power plant processes and suffer from a high degree of simplification.

A chemical absorption process for CO_2 capture based on an aqueous solution of monoethanolamine (MEA) is often considered as the benchmark since commercial processes with MEA that are designed for the application with coal-derived flue gas already exist (cf. e.g., [32, 33]). Consequently, the majority of mentioned publications focus on MEA. There are other well known solvents such as diethanolamine (DEA), methyldiethanolamine (MDEA), and aminomethylpropanol (AMP) for which also a considerable amount of scientific literature is available. Information on novel solvents that have been identified as promising candidates for CO_2 capture from flue gas, however, is scarce and focuses on more fundamental research aspects rather than on the energetic performance of the capture unit or the overall process.

The aim of this work is the energetic evaluation and comparison of chemical solvents for post-combustion CO_2 capture in a typical absorption-desorption process integrated into a coal-fired steam power plant. The performance of the overall process depends on the individual solvent properties in combination with the choice of process parameters of the CO_2 capture unit. A fair comparison on the basis of constant boundary conditions and assumptions therefore requires the optimisation of these parameters for each solvent with respect to a minimal impact on the power plant.

To accomplish this aim, the overall process – including the power plant, the CO_2 capture unit, and the CO_2 compressor – must be represented in adequate detail. The model should be capable of representing any novel chemical solvent for which a minimal amount of information and measurement data is available. To account for the intricate interaction of the CCU with the steam power process and to allow for comprehensive sensitivity analyses of key process parameters, the overall process model needs to be developed in a single simulation environment. To ensure the practicability and relevance of the overall process model, the use of a simulation tool that has proven its applicability in the power generation industry is favourable.