



## **Concurrent flow and countercurrent flow**

Gills are very efficient at taking out what little oxygen is present in the water. This efficiency stems from an adaptation known as countercurrent exchange. Counter current involves blood in the capillaries flowing in the opposite direction to the flow of water flowing over them. difference between the two areas is at its greatest, as compared to when the concentration of a substance differs significantly from one area to another. Consider the animation below the percentage figures refer to the oxygen saturation of the blood or water. In the capillaries of the fish gill, blood with low oxygen concentrations. This creates a steep diffusion gradient along the capillary and favors rapid transfer of oxygen into the blood. Efficiency of the gills is further increased by ventilation, which involves the increase in flow of water over the gills by swimming and by opening and closing their gill flaps. This behaviour draws fresh water into the mouth which passes over the gills. It is common knowledge amongst heat exchanger installation is more efficient than the parallel flow alternative, but for everyone else it can be a bit of a mystery. In this article we will explain what counter flow. What is more efficient than parallel flow. What is counter flow? First we need to understand what the differences are between the flow directions of the hot and cold fluids. With parallel flow the fluids are travelling through the heat exchanger in the same directions. Counter flow installation will have the fluids flowing against each other in opposite directions. Counter flow setup, the fluids are travelling along the heat exchanger in opposite directions. On the diagram above the cold fluid, highlighted in blue, is travelling right to left where as the warm fluid, shown in red and amber, travels left to right. This distributes the heat exchanger and allows for maximum efficiency. In theory, the cold fluid can exit the heat exchanger at a higher temperature than the temperature of the hot fluid outlet, although in reality this is very difficult to achieve. Parallel Flow Heat Exchanger In a parallel flow setup, both the hot fluid and cold fluids are travelling in the same direction as each other. On the diagram above they both flow from right to left. This will still cool the hot fluid down by a considerable amount but is not as efficient as the counter flow system. The benefit; The amount of efficiency gained by using a counter flow system depends on several factors including the flow rates and temperatures (these affect the efficiency gained by using a counter flow system. The benefit; The amount of efficiency gained by using a counter flow system depends on several factors including the flow rates and temperatures (these affect the efficiency gained by using a counter flow system. long) the effect on performance may only be 1 or 2% but with a larger cooler, such as our 2862 model (8" diameter x 1727mm long) the performance can be 10% greater when using a counter flow installation. At Thermex we always design coolers with a counter flow installation. At Thermex we always design coolers with a counter flow installation. information about how to install a heat exchanger can be found in our installation, operation and maintenance manual. For advice or more information about our heat exchangers, please contact us; sales@thermex.co.uk - Richard O'Connor - Thermex Ltd This chapter is related to the aims of Section H3(i) from the 2017 CICM Primary Syllabus, which expects the exam candidate to "describe the principles of dialysis and renal replacement fluid". This having never been examined, one might instead say that realistically it is the bedside nurse, junior colleague or supervisor of training that expects the CICM primary exam candidate to be able to describe these principles. The origin of these notes in their earliest form has its source in the need to answer basic questions from other staff, questions which the author was at that stage unable to field. This chapter explores the dialysis circuit. In summary: Dialysate can be managed in several ways: It remain in a fixed position like a water bath, as in the earliest dialysers It can flow concurrently with blood flow (i.e. in the same direction) It can run counter-current method is the most efficient because it maintains the same concentration gradient along the entire length of the circuit. The difference in efficiency (in terms of urea clearance) is approximately 20% in modern filters, when comparing concurrent arrangements. Why you need a countercurrent arrangements. Why a high concentration of solute and dialysate with a zero concentration of solute would enter the circuit at the same level in the filter. Let's say the blood concentration of the solute is 30mmol/L. As both fluids progress further in the filter, some solute would have been exchanged, and half-way though the filter the concentration in the dialysate is 10mmol/L. Because the concentration gradient is only 10mmol/L. Because the concentration gradient is such an influential factor in determining the solvent diffusive flux, the rate of solute movement into the dialysate is massively affected by this. Let's say that at the end of the circuit the concentration in the blood and in dialysis tubes made by Abel, Rowntree and therefore no diffusion - the maximum removal of the solute has occurred. This is the sort of problem which plagued early experimental dialysis designs. For instance, the fragile colloideon dialysis tubes made by Abel, Rowntree and Turner (the first "proper" dialysis circuit) were bathed in a solution of saline which sat still in a cylindrical tank for the duration of the two hour procedure. The objective of this dialysis was not to cleanse the blood but to collect interesting organic molecules in the dialysis. The clearance of urea by this method was quite slow; little of it was recovered from the fluid. In short, this would not be a satisfactory method of clearing uraemic toxins from the patients. The countercurrent haemodialysis circuit The design of dialysis machines was optimised massively with the advent of the Allwall dialyser in the 1960s, which - instead of a stationary lake of fluid or some sort of rotating drug arrangement- circulated the dialysate fluid over the blood compartment. The effects is best illustrated with a similar diagram: As you can see, the concentration gradient is smaller than with concurrent methods, but it is sustained across the entire filter length. Does this translate into any sort of non-theoretical benefit? Turns out, yes. A group at the Austin hospital in Melbourne (Baldwin et al, 2016) demonstrated with a modern efficient circuit that countercurrent flow of dialysate increases the clearance of urea and creatinine by 20% when compared to a concurrent circuit. The reason for why the magnitude of this difference is so small (i.e. smaller than in the above diagrams is because the movement of even something like urea is very sluggish, and in concurrent flow the concentration of blood urea and dialysate urea would never equilibrate 50:50, In other words, there is always a driving gradient along the entire circuit, even with concurrent flow. Many factors are taken into consideration when designing a rotary kiln. Each affects the ability to produce a desired chemical reaction/phase change in an efficient manner. Characteristics such as bulk density, specific heat, and flowability, among others, all interact to influence how the material will respond to processing, and subsequently, how the kiln will need to be designed around those characteristics to produce the desired result. One critical factor in designing a rotary kiln is the air flow configuration, or the direction in which the process gas flows through the kiln in relation to the material. Rotary kilns are available in two types of air flow configurations: co-current and counter current. Both options have been developed through extensive research and development in order to maximize the thermal efficiency of the process. During the design process, the selection of which air flow configuration will best suit the application is based on the material's properties, as well as overall process requirements. fully understand the benefits each has to offer. Co-Current Air Flow Co-current air flow, which is also referred to as parallel flow, is when the products of combustion flow in the same direction as the material. This immediately puts the coldest material in contact with the hottest gas in the kiln, resulting in a rapid initial temperature change. Co-current air flow, and the same direction as the material in contact with the hottest gas in the kiln, resulting in a rapid initial temperature change. current kilns work best with materials that do not need a gradual temperature increase for a controlled transformation. An organic combustion process commonly uses this air flow configuration, because it does not require a very specific end product. In this example, a waste material (e.g. landfill product) containing both organic and inorganic material is introduced into the kiln. These materials can come into immediate contact with the high heat and the kiln can facilitate the phase change very early on in the process. The organic material is burned off with the high heat and what is left is a dry ash. Counter Current Air Flow Source the phase change very early on in the process. direction of the material flow. In this design, the material is heated gradually while traveling through the kiln. In this configuration, the material comes in contact with the burner being mounted at the end of the thermal processing cycle, less heat is required resulting in decreased fuel consumption. This is illustrated in the tables below. The co-current configuration needs a much higher initial temperature (4000° in this example) to heat the process material from its initial temperature and get the desired phase or chemical change, which in this example, occurs at 2000°. In contrast, in a counter current configuration, the material and the process gas temperature only needs to be slightly higher than the required temperature for the material transformation. The result is a lower burner temperature and lower operating costs. Additionally, the counter current design is commonly used for a more controlled phase or chemical change, where the material temperature needs to be gradually increased to achieve the desired end result. Heat hardening is a common process that utilizes the counter current design is commonly used for a more controlled phase change. The gradual, yet extreme heating process allows for a material such as proppant, to transform into a much harder material. Understanding how each air flow system works is one of the many considerations in designing the most efficient and effective rotary kiln for the job. Both air flow system works is one of the many considerations in designing the most efficient and effective rotary kiln for the job. transformation. FEECO encourages that each material goes through a research and development process at our on-site Innovation Center. The information gained through FEECO's proven testing procedures allows us to design the most efficient and beneficial rotary kiln for our customer's material requirements. Received 15th November 2018, Accepted 8th January 2019First published on 8th January 2019Countercurrent reactors can be utilized in chemical reaction systems which involve either a reacting either a reaction systems which involve eithe the inlet of one participating flow is exposed to the outlet of the opposite flow. A countercurrent configuration can therefore improve the reaction conversion extent and transport properties. Here we formulate a straightforward approach in terms of an exchange coordinate, in order to determine an upper bound of species exchange in such systems, subject to the second law of thermodynamics and conservation of mass. The methodology is independent of the specifics of reactor design and can be generally applied to determine the maximum thermodynamic benefit of using a countercurrent reactor. We then demonstrate the analysis for a number of thermochemical fuel production routes; membrane thermolysis of carbon dioxide, dry methane reforming across a membrane, reverse water gas shift across a membrane, and the thermochemical ceria cycle. Introduction Countercurrent exchange systems are widely applied in industry and frequently observed in nature. For example, a heat exchanger can be arranged in a countercurrent configuration in order to improve overall heat transfer. The same concept is also useful to improve chemical species transfer from one flow to another. A simple example of this occurring in nature is that of gills in fish, which utilize a countercurrent flow arrangement of water and blood to achieve favourable transfer of oxygen.1 In chemical processes with two distinct reacting streams which exchange a species, say A, it may be beneficial to use a countercurrent configuration. This is possible if the reactants have different phases, such as a stream of solid particles reacting with a flow of gas, or bubbles of gas rising against a liquid current. 2 It can also be applied to flows of the same phase if they are separated by an interface such as a species selective membrane, 3 as illustrated in Fig. 1. Despite the common application of such systems, the author couldn't find a standard methodology to determine the thermodynamic limits of countercurrent reactors, either in text books or the literature. A number of models have been developed and applied to specific cases, 4-6 but there is a need for a more generalised approach. Fig. 1 A schematic comparing countercurrent and cocurrent flow, where a species A is exchanged between the two flows. The lack of such a standard methodology has lead many authors to apply simplified models, leading to unphysical results. field of thermochemical fuel production via either membrane reactors or redox cycles.7-12 Thermochemical fuel production systems are proposed as a means of converting heat to chemical energy, by driving chemical reactions that produce a fuel such as syngas. Many authors take the approach of setting the concentration [A] at the exit of each flow to be equal to the concentration at the inlet of the opposite incoming stream. In Fig. 1, this would mean the two lines meet at both ends, which is exchanged. Applying such a model can violate both the second law of thermodynamics and conservation of mass. An analogous error in countercurrent heat exchangers would be to assume that the temperature can be matched at both ends, regardless of the participating flows. This work aims to give researchers a straightforward approach to determine an upper bound on the amount of species exchanged in countercurrent reacting flows. A simple methodology based on a species exchange is developed and used to analyse several examples in thermochemical fuel production systems. The methodology is applied both analysically where an algorithm is outlined for use with thermodynamic software, with links to my implementation made public on GitHub.† The methodology is also developed in a general way and broader in context than the examples discussed. Thermodynamic methodology Consider two distinct streams of matter, which can exchange a species A from one flow to another (flow 1 to flow 2), as illustrated in Fig. 1. For the sake of determining upper bounds we would like to consider a very idealized case, which does not consider any irreversible effects, such as diffusion along the flows. The system is considered to be operating in a steady state, with temperature, pressure, flow rates, species concentration profiles, and heat consumption all assumed to be considered to be in plug flow with no diffusion along the flow. With these assumptions the exchanger can then be considered as a one dimensional interface of length l, along which the species A can be exchanged, as illustrated in Fig. 2. In order to have a spontaneous process with the transfer of species A from flow 1 to flow 2, we must have,  $\mu A_1(x) \ge \mu A_2(x)$ ,  $\forall x \in [0, 1], (1)$  where  $\mu A_1(x) \ge \mu A_2(x)$ ,  $\forall x \in [0, 1], (1)$  where  $\mu A_1(x) \ge \mu A_2(x)$ . coordinate defined with respect to either flow. The flows are separated by an exchange boundary, which could be a phase boundary, which could be a phase boundary or a species selective membrane. It is more convenient to formulated the problem to be independent of the exchange coordinate selective membrane. number of moles of species A that have been exchanged per mole of flow 1 entering the system, by a certain point x along the interface, which is given by (2)where n 1 [mol s-1] is the molar flow 1 to flow 2 as a function of the position, x. Since there is an integral over the length in the numerator, this gives a dimensionless exchange coordinate. For simple systems K will be a monotonic function of x, and the change of co-ordinates is trivial. Eqn (2) then only serves as a formal definition and the system is simply analysed with respect to the exchange co-ordinate K. This gives a much more convenient analysis, as K corresponds directly to changing species number in the flows, and can easily be related to the equilibrium thermodynamics of both flows. We are now interested in finding  $\mu A(\kappa)$  for  $\kappa \in [0, \kappa total]$ , in both flows, where  $\kappa total = \kappa 1(l)$  is the exchange parameter at the exit of the flow (i.e. the total species exchange within the system). With this change of coordinates eqn (1) becomes,  $\mu A, 1(\kappa) \ge \mu A, 2(\kappa)$ .  $\forall \kappa \in [0, \kappa total].(3)$  In words, for any species exchange coordinate  $\kappa$ , the chemical potential of the species A in flow 2. The conservation of mass can be applied to the exchange between the flows, meaning that the number of moles of A to have left flow 1, must be equal to the number of moles to have entered flow 2, at all points along the reactor interface. In a cocurrent system this statement is mathematically trivial and simply means that where ki is the exchange coordinate defined with the integral starting at the inlet of each flow i. For both streams this is equivalent to our definition of the exchange coordinate defined with the integral starting at the inlet of each flow i. For both streams this is equivalent to our definition of the exchange coordinate defined with the integral starting at the inlet of each flow i. For both streams this is equivalent to our definition of the exchange coordinate defined with the integral starting at the inlet of each flow i. For both streams this is equivalent to our definition of the exchange coordinate defined with the integral starting at the inlet of each flow i. For both streams this is equivalent to our definition of the exchange coordinate defined with the integral starting at the inlet of each flow i. For both streams this is equivalent to our definition of the exchange coordinate defined with the integral starting at the integral coordinate above κ. In a countercurrent system we have flow 2 reversed and so conservation of mass means that the exchange coordinate κ can be redefined in flow 2 by, where κ2 is calculated by changing limits of the integral in eqn (2) to be from l to x. This is a simple transformation, where κ2 would be 0 when κ1 = κtotal and vice versa as seen in Fig. 2. Essentially, this reverses flow 2's dependence on the exchange coordinate  $\kappa$ , relative to the cocurrent case, which is illustrated in Fig. 3. Fig. 3 A schematic showing the advantage of countercurrent flow, where the countercurrent flow over cocurrent flow. μA,1 is expected to be a decreasing function of κ, and μA,2 an increasing function, which is illustrated in Fig. 3. This behaviour ensures thermodynamic stability, where any addition of species A to a solution should not decrease μA, and vice versa. Therefore, in a cocurrent system, it is sufficient to obey eqn (3) at the end point of the system κ = κtotal and the thermodynamic upper bound for species exchange ktotal = kmax would be the case where they are equal at the outlet,  $\mu A, 1$  (ktotal). (6) A countercurrent system is not so straightforward. Since flow 2's dependence on  $\kappa$  is reversed (eqn (5)), the chemical potentials  $\mu A, 1$  and  $\mu A, 2$  will both be decreasing functions of  $\kappa$  as illustrated in Fig. 3. This means that with non-linear dependence on  $\kappa$ , it may not be possible to have equal concentrations at ether of the end points, without violating eqn (3) somewhere in the domain  $\kappa \in (0, \kappa \text{total}), (7)$  that they share a common tangent somewhere in between,  $\kappa \in (0, \kappa total)$ , which would satisfy (8) or, it is also possible that all of species A is transferred from flow 1 to flow 2, eqn (3) holds, but neither of the conditions in eqn (7) and (8) are satisfied. In countercurrent reactors it is therefore important to carefully consider the reaction systems of interest. To determine the upper bound of species exchange for a given system one must first fix some parameters such as the temperatures and pressures of the streams. Another key physical parameter which can be set is the relative molar flow rates, which is denoted in this work by ω, (9)For each flow one should then formulate a suitable state function,  $f1(T1,p1,\muA,1,\kappa) = 0$ ,(10)  $f2(T2,p2,\muA,2,\kappa) = 0$ ,(11) which can be used to determine the relation between the chemical potentials and the exchange coordinate  $\mu$ A,i( $\kappa$ ). With the assumption here of plug flow with no diffusion along the flow's and perfect mixing perpendicular, the Gibbs free energy gives such a suitable state function. A simple method of determining the thermodynamic limit on species exchange is then to start with ktotal = 0 and increase this value, until one of the limiting conditions are reached (6 for countercurrent), or as is possible for countercurrent systems, all of the species is transferred. If the flow's are in contact or separated by a thin membrane, most cases will have T1 = T2 and p1 = p2, but the methodology is by no means limited to these cases. For example one could conceivably have a pressure or temperature difference across a membrane, and as long as eqn (3) holds, then we can have spontaneous process with the transfer of species from flow 2. The temperature and pressure could also vary within the system. For example in an adiabatic reactor, where the temperature could also depend on the exchange coordinate T(x) as a result of the heat of the reaction. The examples here are heat driven reactors, which are best approximated as isothermal rather than adiabatic, and so we set both temperatures and pressures to be equal and constant in both flows T1 = T2 = T and p1 = p2 = p. It is important to also understand the context in which can have irreversible effects, such as diffusion along the flow's, and a more sophisticated model would be required to accurately predict performance. With the assumptions used here one can simply set an upper bound on species exchange. It therefore serves as a straightforward check of thermodynamic limits and a means of determining the potential for performance improvement over a cocurrent system. It should also be noted that changes in interface energy have been omitted from the analysis, which in some cases, such as bubble reactors, may play an important role. Examples This section illustrates the analysis of a number of countercurrent reactor systems and makes a brief comparison to previous models and experimental data available in the literature. The examples we consider are, • thermolysis of CO2 with oxygen removal across a membrane, • dry reforming with oxygen exchange across a membrane, • dry reforming with oxygen exchange across a membrane, • ceO2 reduction with a sweep gas removing oxygen, which are illustrated in Fig. 4. Fig. 4 A schematic showing the examples (a-d) in countercurrent configuration. Cases (a) and (d) are solved analytically. Cases (b) and (c) are treated with a more robust numerical method utilizing the thermodynamic library Cantera, 13 with a simple implementation of the methodology in python. + (a) Membrane thermolysis The idea of using a species selective membrane to separate the products of steam thermolysis has been proposed by Fletcher et al. as early as 1977,14,15 with the reaction given by, (12) It is suggested that concentrated solar power could supply the heat for this reaction, and one or both of the products can be selectively removed from the steam using a membrane selective to either hydrogen or oxygen. This direct thermolysis method has been experimentally demonstrated by Tou et al., using a concentrated solar powered reactor to split CO2 with an oxygen selective membrane made from ceria.12 In this system argon acting as an inert sweep gas to carry away oxygen produced by the thermolysis reaction, (13) The heat was supplied using a solar simulator, and the net result was the transfer of oxygen out of the CO2 flow, producing excess CO. Thermodynamic analysis of this countercurrent system utilized a simplified approach of matching the oxygen partial pressure at both inlets and outlets of the reactor, 11,12 which gives unphysical overoptimistic results. Li et al. have provided a more physical model of this system based on a Gibbs critereon dGT,  $p \le 0.30$  which is in agreement with the results presented here. A schematic of the system can be seen in Fig. 4(a), which can be modeled according to the methodology described in the previous section to determine the thermodynamic limits. Note that the analysis presented would be identical if the CO2 were replaced with steam for H2O thermolysis. The system is modeled as isothermal at a temperature of 1 bar, which allows for direct comparison of our model to the experimental work of Tou et al.12 Since oxygen is being exchanged between the two flows, we can define our exchange coordinate as (14)In this case  $\kappa = 0.5$  would correspond to complete transfer of the sweep gas to the CO2 is used as a free control parameter, (15) The thermodynamics of both flows can be well approximated as ideal gas solutions giving (16)Both flows have equal temperature, and so we can use the oxygen partial pressure instead of the chemical potential in egn (3) giving the condition,  $pO2.1(\kappa) \ge pO2.2(\kappa)$ .  $\forall \kappa \in [0, \kappa total](17)$ We can also use the partial pressure to check the conditions given in egn (6)-(8). We need to determine  $pO2.2(\kappa)$ .  $\forall \kappa \in [0, \kappa total](17)$ We can also use the partial pressure to check the conditions given in egn (6)-(8). we have these functions we can apply the methodology developed and determine the maximum exchange extent kmax for a given temperature pressure and relative flow rates. For the sweep gas determining pO2(k) is straightforward. Assuming we have a sweep gas determining pO2(k) is straightforward. the sweep gas stream (flow 2) is given by (18) The derivative of this function is then given by, (19) In the countercurrent case we use the substitution  $\kappa' = \kappa \text{total} - \kappa$  in eqn (18) and (19). In the countercurrent case we use the substitution  $\kappa' = \kappa \text{total} - \kappa$  in eqn (18) and (19). In the countercurrent case we use the substitution  $\kappa' = \kappa \text{total} - \kappa$  in eqn (18) and (19). In the countercurrent case we use the substitution  $\kappa' = \kappa \text{total} - \kappa$  in eqn (18) and (19). In the countercurrent case we use the substitution  $\kappa' = \kappa \text{total} - \kappa$  in eqn (18) and (19). described by the variance in the Gibbs free energy, (20) which should be zero at equilibrium. This assumes the formation of other species (e.g. atomic oxygen) is negligible and the partial pressures are related by, pCO + pCO2 + pO2 = 1.(21)Eqn (20) and (21) can be solved to get pCO and pCO2 for a given pO2, (22) pCO2(pO2) = p - pCO(pO2) - pO2, (20) pCO2 + pCO(23) where is the equilibrium constant. The exchange coordinate in the CO2 flow can be formulated in terms of the partial pressures as, (24) where the first term is the total extent of oxygen formation in the splitting reaction, and the second term accounts for the oxygen gas which remains in flow 1. Subbing eqn (22) and (23) into eqn (24) gives (25) which can then be inverted to get pO2,1(k), although this does not yield a simple analytical expression. For that reason numerical solutions of eqn (25) for a given k were used. For the derivative we can use the calculus identity (26) to determine . The thermodynamic limiting case can then be found, by starting from ktotal = 0, and increasing this value until our stop conditions given in eqn (6)-(8) are reached, corresponding to ktotal = kmax. An example of the limiting case is illustrated in Fig. 5(a), for both cocurrent and countercurrent flow configurations. In the cocurrent case the sweep gas flow (CC) has an increasing dependence on k and the partial pressures of the two flows meet at the maximum exchange extent, satisfying eqn (6). In the countercurrent case however both the CO2 flow and the sweep gas flow (CT) have a decreasing dependence on  $\kappa$ , and they share a common tangent satisfying the conditions given in eqn (8). Fig. 5 (a) Plots of oxygen partial pressure vs. exchange extent  $\kappa$  for the CO2 stream given by eqn (25), cocurrent sweep gas flow CC given by eqn (18), and countercurrent sweep gas flow CT given by eqn (18) with  $\kappa' = \kappa \max - \kappa$ . The initial oxygen partial pressure in the sweep gas  $\phi$  and the kmax in both cases are also labeled. (b) Mole fraction of CO in the CO2 stream plotted for cocurrent and countercurrent flow configurations at the same conditions listed in (a). Also shown is an experimental a point corresponding to Tou et al.'s experimental demonstration of this system in countercurrent configuration.12 Fig. 5(b) shows the dependence of the mole fraction of CO in the product stream, on the relative flow rate ω. This is equivalent to the CO2 conversion, where a value of one would indicate complete conversion. It can be seen that the countercurrent arrangement almost doubles the formation of CO (and kmax) relative to the cocurrent exchange of a cocurrent systems. In general, the conversion of CO2 to CO increases with increasing  $\omega$ , and approaches a thermodynamic limit which is determined by the oxygen impurity in the sweep gas. The sweep gas impurity was selected to match conditions reported by Tou et al., where we have also included the conversion extent measured for their countercurrent reactor.12 The experimental value lies above the cocurrent model (CC), indicating that there was a real benefit to countercurrent operation. It also lies below the countercurrent thermodynamic limit, which it should. Using the model applied by previous authors of matching the partial pressures at both the entrance and exit of the countercurrent thermodynamic limit, which it should. on the purity of the sweep gas.11,12 This is a counter intuitive result, where a fully pure sweep gas would then offer complete conversion. It can be seen by the differing shapes of the curves in Fig. 5(a), that matching both ends in the countercurrent case would not be possible, without violating the conservation of mass and/or the second law of thermodynamics (eqn (17)). Correctly analysing the benefit of countercurrent operation for such a reactor shows that the conversion extent of CO2 (or H2O) at 1500 °C will be very small, unless huge quantities of very pure sweep gas are fed to the reactor. The thermodynamics of this membrane reactor system for thermolysis of CO2 or H2O, indicate that very high temperatures and/or very low oxygen partial pressures are required to achieve significant conversion of the reactants. This is unlikely to offer a practical or economically competitive means of converting heat to chemical energy. (b) Membrane reforming In the above section we used an analytical approach to solve a simple membrane countercurrent problem. In this case we look at a more complicated reaction system, and apply the methodology developed with a robust numerical analysis of the thermodynamic equilibrium. An interesting variation on the above process is to use methane partial oxidation taking place in flow 2, (27) This system is illustrated in Fig. 4(b), where CO2 and CH4 flows are separated by an oxygen selective membrane. If CH4 and CO2 are supplied in equal stoichiometry, the sum of the reactions on both sides is then dry reforming (28) which consumes heat and gives two product streams, CO and 2H2:1CO syngas. This type of membrane methane reforming has been demonstrated by several authors experimentally.3,16,17 For methane partial oxidation, there can also be the formation of CO2 and H2O, and so a simplified analytical approach will not suffice. Instead the thermodynamics of the reactions was modeled using the software Cantera,13 and it's gri30 database, which contains all of the relevant species. This software uses an element potential method to equilibrate an initially defined mixture of gases by minimizing the Gibbs free energy for the system.18 Since the same reaction is taking place in flow 1 as the previous example, our exchange parameter can again be defined by eqn (14), where we do not consider (or indeed expect) the reduction of CO to carbon, so that  $\kappa = 0.5$  represents complete conversion. Here we consider the case with equal flow rates. (29) at a pressure of 1 bar in both streams, and study the equilibrium limitations as a function of temperature. To apply the methodology we need to determine the oxygen chemical potential or  $pO2(\kappa)$  in both streams. For the CO2 stream this can be achieved by taking an initial mixture of CO and O2 corresponding to a given κ, (30) and finding the thermodynamic equilibrium composition as the CO2 splitting reaction for a given κ as it is simply the reverse reaction. Similarly for the methane partial oxidation we consider an initial mixture of methane and find the thermodynamic equilibrium composition, (31)to determine pO2,2(κ). numerically checking if the condition in eqn (6) holds for the cocurrent case, and if eqn (3) holds for all x in the countercurrent case. This algorithm is graphically illustrated in Fig. 6, where it is important to reverse the order of the countercurrent case. This algorithm is graphically illustrated in Fig. 6, where it is important to reverse the order of the countercurrent case. maximum possible oxygen exchange in a dry reforming membrane reactor. This was implemented in Python using Cantera, and the code has been made publicly available. Fig. 7(a) shows an example of the equilibrium partial pressure profiles with respect to the exchange coordinate  $\kappa$ . Here it can be seen that the CO2 oxygen release profile and CH4 oxygen uptake profile in countercurrent configuration allow for complete exchange (kmax,CT = 0.5) and complete CO2 conversion to CO, even at 600 °C. The cocurrent case on the other hand only exchanges about two thirds of the oxygen. Fig. 7 (a) Plots of oxygen partial pressure vs. exchange extent k for the CO2 stream, the cocurrent CH4 flow CC, and countercurrent CH4 flow CT. (b) CO2 conversion, plotted as a function of the temperature for cocurrent with also consider CH4 conversion, as further oxidation of CO and H2 to CO2 and H2O in flow 2, can decrease the conversion of methane to syngas. The methane conversion can be determined from the mole fractions using the carbon balance, (32) which is plotted along with the CO2 conversion in Fig. 7(b). Here it can be seen that although the conversion for CO2 conversion in Fig. 7(b). conversion at low temperatures, both countercurrent and cocurrent configurations require high temperatures of 800 °C plus, to achieve a high methane conversion and syngas production in flow 2. The thermodynamic benefits of countercurrent are therefore mostly limited to the CO2 conversion. In the literature the results for conversion extents by Michalsky et al., fall well short of the thermodynamic limit,3 indicating that the system may have been kinetically limited. This is supported by the work of Jin et al., who used a catalyst in their membrane reactor and achieved higher conversions.16 The reactor of Jin et al., who used a catalyst in their membrane reactor and achieved higher conversions.16 The reactor of Jin et al., who used a catalyst in their membrane reactor and achieved higher conversions.16 The reactor of Jin et al., who used a catalyst in their membrane reactor and achieved higher conversions.16 The reactor of Jin et al., who used a catalyst in their membrane reactor and achieved higher conversions.16 The reactor of Jin et al., who used a catalyst in their membrane reactor and achieved higher conversions.16 The reactor of Jin et al., who used a catalyst in their membrane reactor and achieved higher conversions.16 The reactor of Jin et al., who used a catalyst in their membrane reactor and achieved higher conversions.16 The reactor of Jin et al., who used a catalyst in their membrane reactor and achieved higher conversions.16 The reactor of Jin et al., who used a catalyst in their membrane reactor and achieved higher conversions.16 The reactor of Jin et al., who used a catalyst in the system may have been kinetically limited. 3CO2:1CH4, i.e.  $\omega = 0.33$ . The conversion extent trends were similar to the thermodynamic analysis presented here, with a steeper dependence on temperatures. Unfortunately, in these demonstrations the reactants are diluted in inert gases for experimental analysis purposes, which makes a more quantitative comparison difficult. (c) Membrane reverse water gas shift It is also interesting to consider other gases than CH4 to reduce CO2 in a membrane reactor. (33)We then have CO2 and H2 flows separated by an oxygen selective membrane. The sum of the reactions on both sides of the membrane is the reverse water gas shift (RWGS) (34) which consumes a small amount of heat and with a membrane reactor gives two product streams, CO/CO2 and H2/H2O. This reaction is of industrial relevance for producing syngas (H2 + CO mixtures) from a hydrogen source. Syngas is a highly valuable product used in many industrial processes, including gas to liquids plants for producing fuels. This is of particular interest if combined with renewable sources of hydrogen, allowing for the storage of renewable energy sources in highly valuable liquid fuels. The reverse water gas shift however typically requires very high temperatures (>800 °C), making industrial implementation a challenge. Some concepts have been considered to reduce the temperature, such as using a steam absorbent to shift the equilibrium to higher conversion.19 The analysis here shows that using a countercurrent membrane reactor with separate H2 and CO2 streams, has promising thermodynamics for relatively low temperature operation. Taking renewable hydrogen followed by methanol synthesis as an example application, one could produce a suitable syngas by feeding three times as much hydrogen as CO2, (35)The resulting steam would then be condensed out of the hydrogen as CO2, (35)The resulting steam would then be condensed out of the hydrogen as CO2, (35)The resulting steam would then be condensed out of the hydrogen as CO2, (35)The resulting steam would then be condensed out of the hydrogen as CO2, (35)The resulting steam would be mixed and then fed to the methanol synthesis process. The feed ratio of 3:1 ensures that we will have a syngas composition suitable for methanol synthesis even without complete conversion of the CO2 according to the reactions,  $3H2 + CO2 \rightarrow CH3OH + H2O.(37)$ The reactor is modelled to operate at 1 bar, but since the RWGS reaction does not change the number of moles of gas, the thermodynamic conversion is independent of pressure. Since the same reaction is taking place in flow 1 as the previous two examples, our exchange parameter is again given by eqn (14). The thermodynamic library Cantera is used to model thermodynamics of each stream. The CO2 stream was modelled according to eqn (30), to determine the oxygen partial pressure pO2,1(κ). Similarly for the H2 stream the equilibrium is calculated according to, (38)to determine pO2,2(κ). The same numerical algorithm given shown in Fig. 6 is used to find the thermodynamic limits. Fig. 8(a) shows an example of the partial pressure profiles with respect to the exchange coordinate κ. Here it can be seen that the CO2 oxygen release profile and H2 flow's oxygen uptake profile in countercurrent case. In the countercurrent case, more than 90% of the oxygen is exchanged corresponding to almost double the oxygen exchange as the cocurrent case. In the countercurrent case, more than 90% of the oxygen is exchanged corresponding to almost double the oxygen partial pressure vs. exchange extent κ for the CO2 stream, the cocurrent H2 flow CC, and countercurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of the temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of temperature for cocurrent H2 flow CT. (b) Maximum CO2 conversion (=2κmax) in flow 1 plotted as a function of temperature for cocurrent H2 flow CT. (b) Maximum CO2 conv shows the conversion extent of CO2 for both cases, and for comparison an equilibrium calculation of the standard RWGS reaction with 3H2 + CO2. As one would intuitively expect the thermodynamic conversion limit in the cocurrent case is identical to that of the standard RWGS process. The analysis shows that a countercurrent membrane reactor has promise for a low temperature reverse water gas shift process. The author could not find any experimental work or otherwise on this study. A physical implementation of this system have kinetic issues at low temperatures (

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