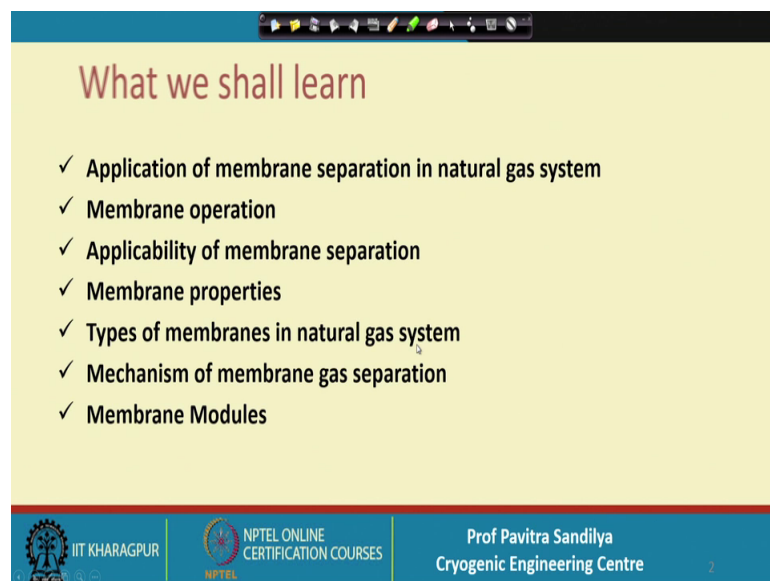


Upstream LNG Technology
Prof. Pavitra Sandilya
Department of Cryogenic Engineering Centre
Indian Institute of Technology, Kharagpur

Lecture – 34
Membrane separation in natural gas systems



Welcome, now we shall be learning about another type of separation processes used in the natural gas industries that is the Membrane separation.

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What we shall learn

- ✓ Application of membrane separation in natural gas system
- ✓ Membrane operation
- ✓ Applicability of membrane separation
- ✓ Membrane properties
- ✓ Types of membranes in natural gas system
- ✓ Mechanism of membrane gas separation
- ✓ Membrane Modules

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In this particular lecture we shall be learning about the application of the membrane separation in natural gas systems, then the different types of membrane operations, the applicability of the membrane operation we shall look into is advantages these advantages then membrane properties, and the types of membrane used in the natural gas systems, then mechanism of membrane gas separation and the different types of membrane modules that may be used for the natural gas systems. These are the just some overviews of membrane systems the detailed lectures will not be given in this particular thing.

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Application of membrane separation in natural gas system

- ✓ Dehydration of natural gas
- ✓ Acid gas removal
 - Carbon dioxide
 - Hydrogen sulphide etc.
- ✓ Nitrogen removal

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So, here we have some applications of the membrane separation for the natural gas processing; one is the dehydration of natural gas, then acid gas removal like carbon dioxide, hydrogen sulfide and then we have the removal of nitrogen.

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Membrane

- ✓ Is a semi-permeable (or permselective) barrier that allows selective permeation of certain species through it.
 - May be used for both liquid and gas separation

Liquid separation by microfiltration, ultrafiltration (UF), reverse osmosis (RO), dialysis etc.

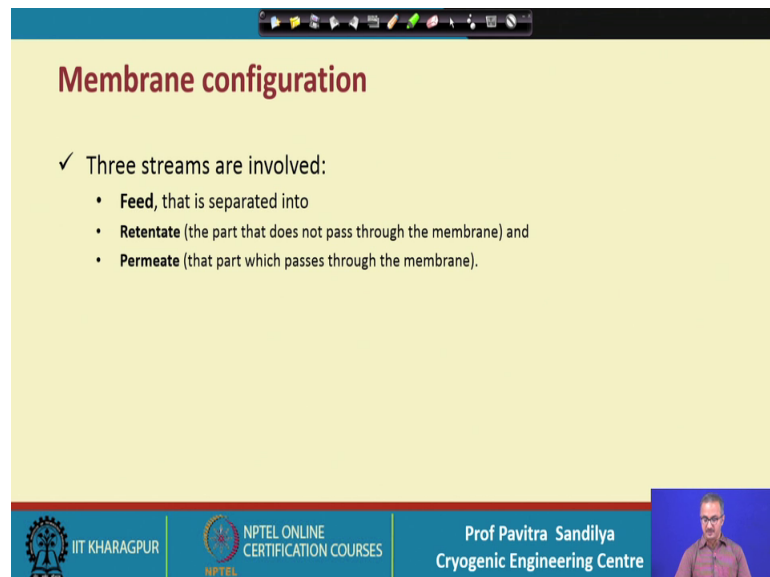
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First let us define what is a membrane. A membrane is a semi-permeable or what is called a permselective barrier that allows the selective permeation of certain species through it. Now permselective come from two words permeation plus selective; that means, what a membrane does that it allows some of the components to pass through it,

and it does not allow some of the components to pass through it that is how it is able to effect separation.

So, this membrane separation is used both for liquid separation, gas separation and we are using the membrane separation at our household application like in the water a purification we use at our home and here we you might have heard some of these names like microfiltration, ultrafiltration, reverse osmosis etcetera which are used in the water purification at our home, but we shall not be talking about liquid separation by membrane we shall be focusing on only the gas separation

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Membrane configuration

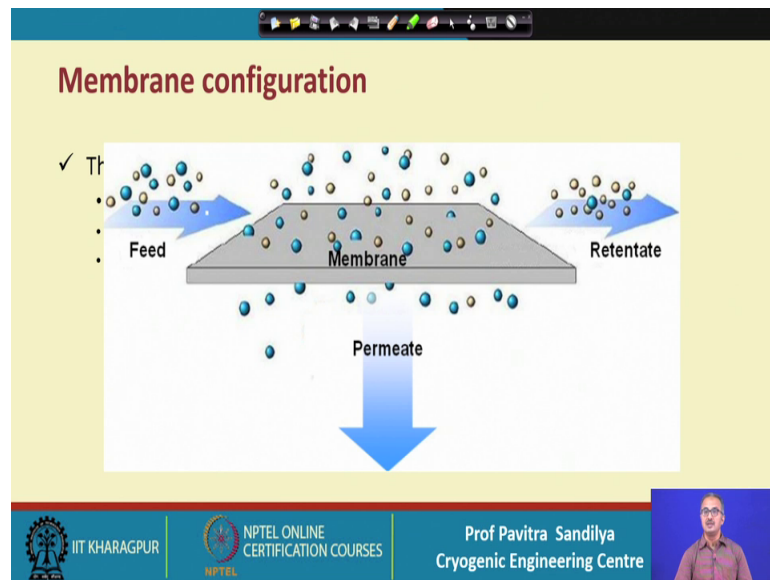
✓ Three streams are involved:

- **Feed**, that is separated into
- **Retentate** (the part that does not pass through the membrane) and
- **Permeate** (that part which passes through the membrane).

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Now let us look at a typical membrane configuration. In the membrane configuration we have three streams one is the feed the feed is separated into a retentate; that means, the part of the total feed mixture, that is retained by the membrane and another is the permeate and this is that part of the feed that goes through the membrane.

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Then here is a pictorial representation of the configuration, here we find we have the feed which is going coming here, and the feed is represented by these circles of various colors.

And we find the feed when it passes over the membrane some of these permeate through and here in this case we find that this blue color balls are coming through and in between we have some other golden color balls; that means, this permeate is primarily the one of the components and rest of the components are passing over the membrane and this is coming out and this is called a retentate that is the fraction which is retained by the membrane. So, this is a typical representation of the membrane configuration.

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Advantages of membrane separation

- Light weight (modular construction)
- Large turndown ratio (ratio of maximum to minimum capacity)
- Low maintenance
- Low operating cost

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Next we come to the applicability and let us look into the advantages and disadvantages; here we have that the membrane is advantages that it is quite lightweight because these come in modular construction, we have small modules of the membranes and we have large turndown ratio that is we the ratio of the maximum to the minimum capacity and this also is because that when we want to increase the capacity, we may increase the number of modules and then the cost and the maintenance are also quite low. So, there that way we find that the membrane give some advantage for the cost reduction.

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Disadvantages of membrane separation

- Proves expensive if multi-staging and/or recycling is done.
- Membranes are prone to
 - Mechanical and thermal damage at high pressures and high temperatures
 - Fouling, and deposition of particulates(→ needs feed pre-treatment)
- In case of natural gas, some methane is lost with water vapor permeating the membrane, making membrane process noncompetitive with absorption.

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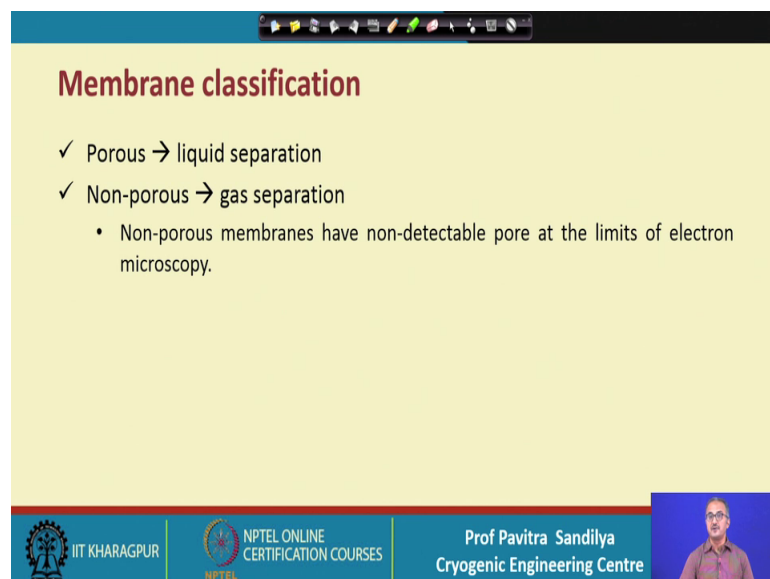
On the other hand it has some disadvantages and these are that it proves expensive if we need some multi staging that is we need several modules and we need to recycle we shall look into this kind of configurations later, but if you are going for multi staging or recycling, the cost increases.

The membranes are prone to mechanical and thermal damages at high pressure and high temperatures and there are also prone to fouling; fouling means the deposition on some unwanted products on the membrane surface and if the fouling occurs what will happen that the, the feed the permeate or the various types of the molecules will not be able to pass through the membrane properly, and a resistance to the separation will increase and that will decrease the efficiency of separation.

So, membranes are prone to fouling and deposit of particulate matters and if there are particulate matters in the membrane, then these particulates need to be separated by some pretreatment method before sending the feed on the membrane surface.

For natural gas systems it is found that, some of the methane also passes because when we talk of dehydration, we want the water to pass through the membrane whereas, the methyl should not be passing through, but we find that some of the methane also goes to the membrane and that we what we find that we are losing some of the methane from the natural gas during the dehydration of natural gas by membrane, and this makes it a noncompetitive with respect to the absorption process.

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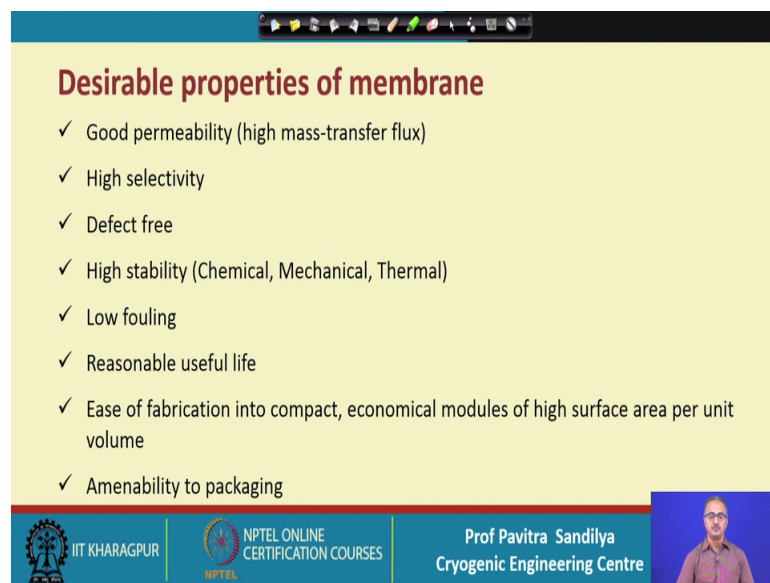
Membrane classification

- ✓ Porous → liquid separation
- ✓ Non-porous → gas separation
 - Non-porous membranes have non-detectable pore at the limits of electron microscopy.

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Now, there are various ways of classifying the membrane, we have porous membrane and non porous membranes. Porous membranes are used when the molecular size is more and non porous are used when the molecular sizes are less and porous membranes are used for the liquid separation and the non porous for gas separation, we shall be looking into only the gas separation that is a non porous type of membranes. And when is a non porous what it means is that, we have different we have pores, but these pores are non-detectable by the limits of the electron microscopy. So, it is not totally non porous, but we are the pore size are very small.

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The slide displays a list of desirable properties for a membrane. The title is 'Desirable properties of membrane'. The list includes: Good permeability (high mass-transfer flux), High selectivity, Defect free, High stability (Chemical, Mechanical, Thermal), Low fouling, Reasonable useful life, Ease of fabrication into compact, economical modules of high surface area per unit volume, and Amenability to packaging. The slide footer contains logos for IIT KHARAGPUR, NPTEL ONLINE CERTIFICATION COURSES, and Prof Pavitra Sandilya from the Cryogenic Engineering Centre.

- ✓ Good permeability (high mass-transfer flux)
- ✓ High selectivity
- ✓ Defect free
- ✓ High stability (Chemical, Mechanical, Thermal)
- ✓ Low fouling
- ✓ Reasonable useful life
- ✓ Ease of fabrication into compact, economical modules of high surface area per unit volume
- ✓ Amenability to packaging

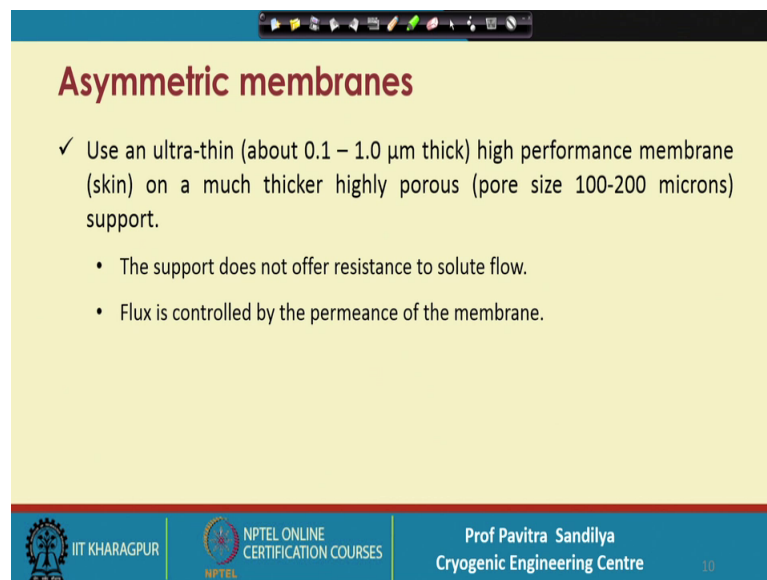
So, these are some of the desirable properties of the membrane, one is that it should have good permeability and if it has good permeability it means the mass flux will be high, then you should be selective; that means, it should be selectively permeating some of the solutes to pass through it so that we get the separation and then it should be defect free; that means, there should not be any kind of pinhole in the membrane and there should not be any distortion so, that we can retain the same efficacy of separation throughout the membrane module.

Then we have high stability; the stability comes with respect to chemical, mechanical and thermal stability. Chemical means it should not get dissolved or it should not react with any of the components, mechanical means it should be able to withstand the high

pressure difference that occurs across the membrane surface and thermal means it should not get damaged by any high temperature.

Then we have low fouling tendency; that means, it should not be retaining any of the waste matters or fouling matters on it, then you should have reasonable useful life. Then also it should be easy to fabricate because if we have ease of fabrication, then we can make a very compact model which would be economic and we can also at the same time have high surface area per unit volume of the membrane. And lastly we have the amenability to packaging; that means, we should be able to package the whole module easily.

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Asymmetric membranes

- ✓ Use an ultra-thin (about 0.1 – 1.0 μm thick) high performance membrane (skin) on a much thicker highly porous (pore size 100-200 microns) support.
 - The support does not offer resistance to solute flow.
 - Flux is controlled by the permeance of the membrane.

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Now, let us come to the membrane which is used for the gas separation and in this we have the asymmetric membranes, what is asymmetric? It means that we have a ultra thin it is about 0.1 to 1 micron thick high performance membrane and this particular ultra thin membrane is called the skin on a much thicker in highly porous. Highly porous means 100 to 200 microns support; that means, if I have the only skin, the skin will not be having enough mechanical stability it may buckle.

So, to prevent this buckling of the thin membrane the skin what we need? We need a support and the support is made quite thick, but we want that the support should not be offering any kind of resistance to the permeation of the solute. So, that is why the pore size of the supports are much bigger in comparison to the pore sizes of the skin.

So, this pore and that is how we are trying to ensure that the supports are not giving any kind of resistance to the mass transfer. So, we have the thin skin that is supported by a thick support and that is why it is called asymmetric membrane. Now the support should not be offering the resistance to flow and the flux is controlled by the membrane surface.

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Asymmetric membranes

- ✓ Use an ultra-thin (about 0.1 – 1.0 μm thick) high performance membrane (skin) supported by a porous support system.
- The skin layer is almost pore-free.
- Flux is controlled by the skin layer.

The diagram illustrates the structure of an asymmetric membrane. It consists of a thin, dense skin layer on top of a thick, porous support layer. A circular inset provides a magnified view of the skin layer, showing its dense, non-porous structure. The porous support layer is shown with large, interconnected pores. Labels 'Skin Layer' and 'Porous Support' point to their respective parts. The slide footer includes the IIT Kharagpur logo, the NPTEL logo, and the text 'NPTEL ONLINE CERTIFICATION COURSES', 'Prof Pavitra Sandilya', 'Cryogenic Engineering Centre', and the number '10'.

So, here is a typical configuration of the asymmetric membrane, here we find that we have a porous support on which we are supporting the skin layer and here we are showing that how this supports, here we are showing the skin is almost there is no pores and with comparison to the skin we have that big pores of the support system so; that means, the rate of permeation is dictated by the skin and not by the support.

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Membranes for natural gas separation

- ✓ Polymeric membranes
 - Polyimide (CO₂ removal)
 - Cellulose acetate (CO₂ removal)
 - Perfluoro polymer (CO₂, N₂, C₃₊ hydrocarbons removal)
 - Silicone rubber (CO₂, N₂, C₃₊ hydrocarbons removal)
 - Polysulfone (water removal)

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So, in case of natural gas separation, we use polymeric membranes and here we are showing some of the polymeric membranes polyimide for carbon dioxide removal then cellulose acetate, a perfluoro polymer, silicon rubber and polysulfone and we are we have shown the various applications of these membranes in the natural gas processing.

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Permeation mechanism through a non-porous membrane

- ✓ Solution-diffusion mechanism
 - Dissolution of the gas into the high-pressure upstream face of the membrane
 - Diffusion of the gas through the membrane
 - Desorption from the low-pressure downstream face of the membrane

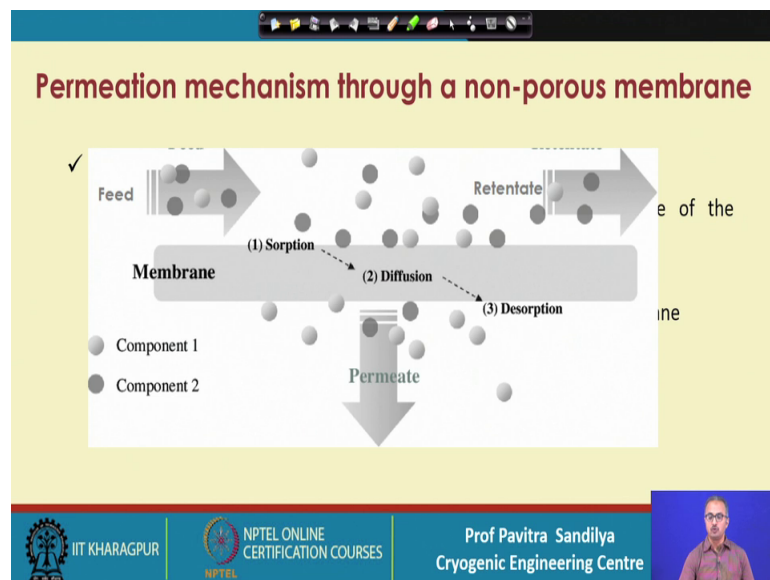
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Now, let us look into the mechanism of separation through these non porous membranes. The mechanism is called solution diffusion mechanism and in this by solution remains the gas will be dissolving at the high pressure upstream. This dissolution is basically a

process of adsorption, it is not by dissolving the way sugar dissolves in water not that way, but it is basically a an adsorption process so that is because dissolution. So, that the gas has to be first dissolved has to dissolve on the upstream side and on the upstream side we have a high pressure, on the downstream side we have a low pressure and this pressure difference across the membrane surface is the driving force for the solute movement.

So, after dissolution of the gas on the upstream side which is at high pressure, then the gas will diffuse through the membrane and after reaching the permeate side, it has to get desorbed from the downstream low pressure side of the membrane.

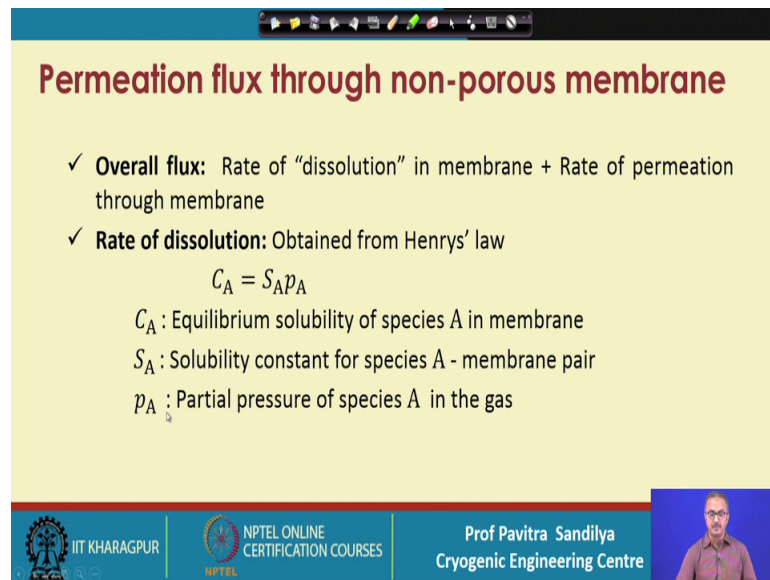
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And here we show pictorially the whole mechanism, that the feed is coming we have two separate colored balls to show the mixture, and this feed is coming and then some of these balls are getting absorbed or dissolved on the upstream side.

And then they are diffusing through the membrane and ultimately when they reach the permeate side, they get desorbed here and we find in this case, the grey color balls this are the coming on the more on the permeate side, where the dark colored balls are coming it being retained on the feed side and some of them are also permeating through the membrane. So, that is how we are able to separate a gas, but this by this solution diffusion mechanism.

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Permeation flux through non-porous membrane

- ✓ **Overall flux:** Rate of “dissolution” in membrane + Rate of permeation through membrane
- ✓ **Rate of dissolution:** Obtained from Henrys’ law

$$C_A = S_A p_A$$

C_A : Equilibrium solubility of species A in membrane
 S_A : Solubility constant for species A - membrane pair
 p_A : Partial pressure of species A in the gas

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Now, here we have to find out the overall flux of the particular solute, we have to consider the rate of dissolution in the membrane and rate of permeation. So, this is how we are getting rate of dissolution, we are using Henrys law, in which we have this C_A equal to $S_A p_A$. C_A is defined as the equilibrium solubility of species A in the membrane, S is the solubility constant for species a membrane pair and p is the partial pressure of species A in the gas on the permeate side the on the sorry on the feed side. Please understand this that this S_A depends on the pairing of the species and the membrane; that means, for different pairs of the species and the membrane we shall be having different values of the S_A . It is something what we have learnt in case of adsorption that in adsorption also the rate of adsorption depends on the solute adsorbent pair.

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Permeation flux through non-porous membrane

✓ Rate of permeation

$$J_A = \frac{\bar{P}_A (p_{A1} - p_{A2})}{l_m}$$

p_{A1} and p_{A2} : Partial pressures of a component A in permeate and retentate
 l_m : Thickness of the membrane
 \bar{P}_A : Permeability of A ($\bar{P}_A = D_A S_A$)
 D_A : Diffusivity of A through the membrane

✓ Common unit of permeability: **barrer**.

• 1 barrer = $10^{-10} \frac{\text{cm}^3 (\text{STP})\text{cm}}{(\text{cm}^2 \text{ membrane})(\text{cm Hg})(\text{s})}$

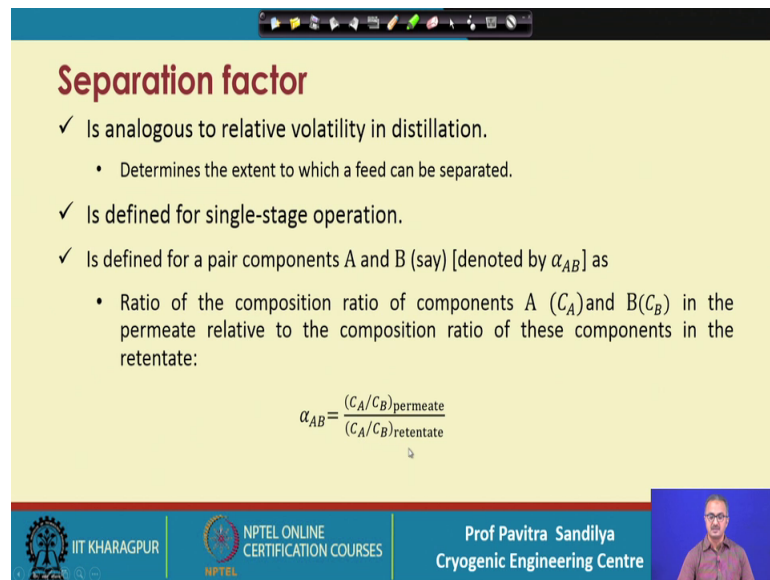
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So, here we are the expression to find out the rate of permeation, in which we are finding the driving force is the difference in the partial pressure of the species A and 1 and 2 represent the feed side and the permeate side respectively. As long as the feed side partial pressure is more than the feed partial pressure on the permeate side, we shall be having this particular solute transfer. Please understand this the driving force is not the total pressure, but the partial pressure of the particular component.

And l_m is the thickness of the membrane whereas, P is the permeability and D is the diffusivity which is used to calculate the permeability of the species through the membrane. Now if we put this particular equation in electrical analogy we find J_A corresponds to the current. This difference in the partial pressures corresponds to the voltage then in that case l_m by P_A will represent the resistance of to the mass flux of the particular species through the membrane.

The unit of permeability is barrier and the barrier is defined like this, that it is 10^{-10} to the power minus 10 cubic centimeter STP, centimeter this centimeter represents the thickness of the membrane, then square centimeter of the membrane surface then this is the pressure partial pressure in terms of centimeter of mercury, and this is per second. So, this is how the barrier is defined.

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Separation factor

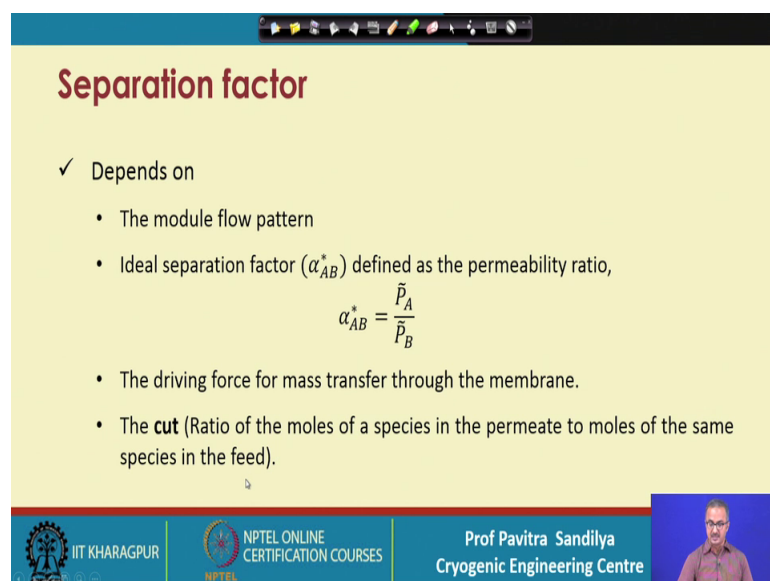
- ✓ Is analogous to relative volatility in distillation.
 - Determines the extent to which a feed can be separated.
- ✓ Is defined for single-stage operation.
- ✓ Is defined for a pair components A and B (say) [denoted by α_{AB}] as
 - Ratio of the composition ratio of components A (C_A) and B (C_B) in the permeate relative to the composition ratio of these components in the retentate:

$$\alpha_{AB} = \frac{(C_A/C_B)_{\text{permeate}}}{(C_A/C_B)_{\text{retentate}}}$$

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Now, we have the separation factor, which represents it is something analogous to the relative volatility, it determines the extent to which the feed can be separated and is defined for a single stage operation, for a pair of components A and B denoted by alpha AB, it is defined as the ratio of the composition of a CA and the composition BCB in the permeate relative to the same ratio on the feed side or the retentate side, and here we have the definition of alpha AB that CA by CB on the permeate side to CA by C B the retentate side this is how we define the separation factor.

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Separation factor

- ✓ Depends on
 - The module flow pattern
 - Ideal separation factor (α_{AB}^*) defined as the permeability ratio,

$$\alpha_{AB}^* = \frac{\tilde{P}_A}{\tilde{P}_B}$$

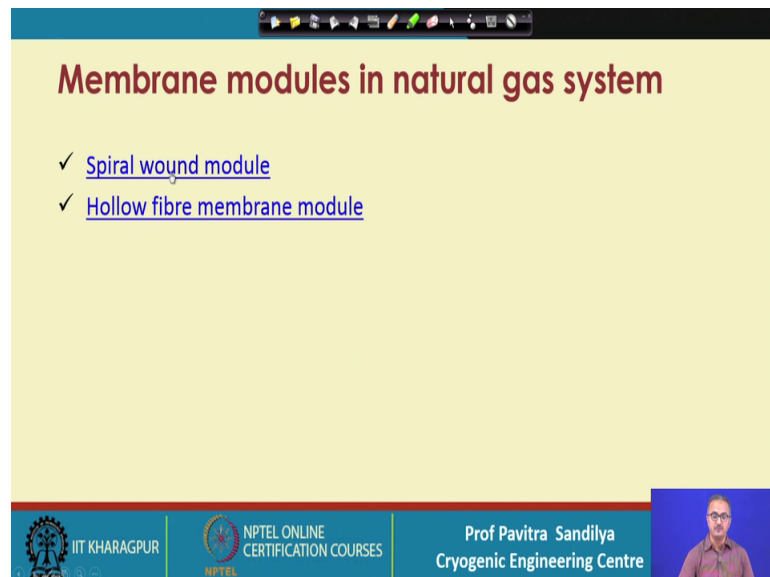
- The driving force for mass transfer through the membrane.
- The **cut** (Ratio of the moles of a species in the permeate to moles of the same species in the feed).

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And it depends on the flow pattern of the in the module about which we shall see a bit later, and then the ideal separation factor that is α_{AB}^* , which is defined as the ratio of the permeabilities and here it is the α_{AB}^* is equal to the permeability of component A to permeability component B.

So, we can see that α_{BA} will be the α_{BA}^* will be the inverse of α_{AB}^* . And the driving force of the mass transfer through the membrane that is the when Δp and the cut is defined as the mole of a particular species in the permeate to the mole of the same particular species in the retentate.

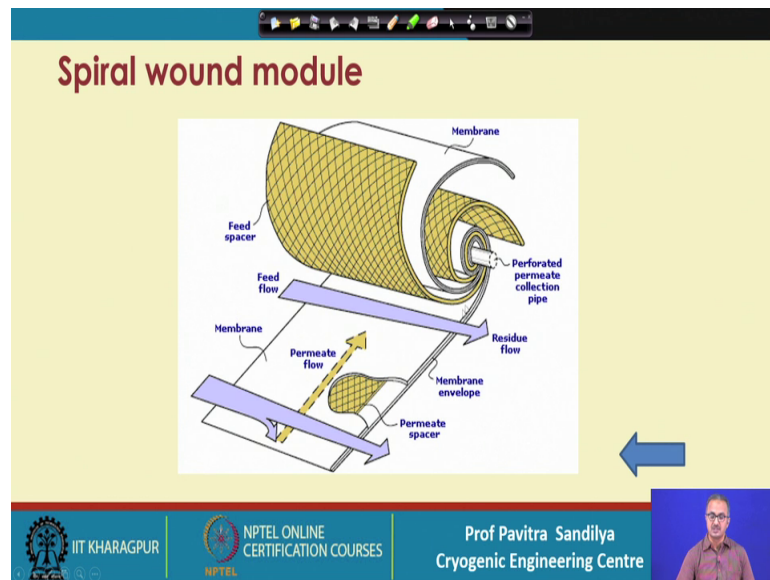
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The slide is titled "Membrane modules in natural gas system" in a dark red font. Below the title, there are two bullet points, each with a checkmark and a blue underlined link: "✓ [Spiral woynd module](#)" and "✓ [Hollow fibre membrane module](#)". The slide has a yellow background. At the bottom, there is a blue footer bar containing the IIT Kharagpur logo, the NPTEL Online Certification Courses logo, the name "Prof Pavitra Sandilya" and "Cryogenic Engineering Centre", and a small video inset of the professor.

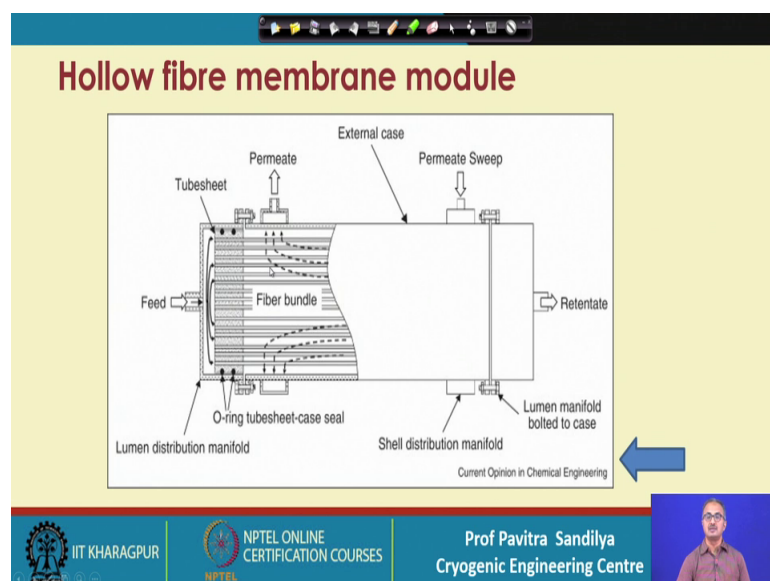
So, here we have different types of membrane modules and we are talking about the typical membrane modules, which are used for the natural gas systems in this case we have two types of membrane modules. So, first is a spiral wound and as the hollow fiber.

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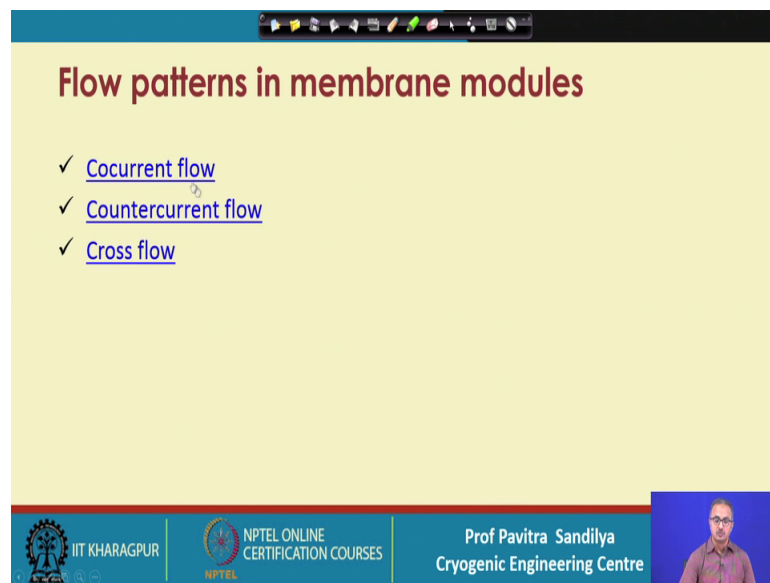
What is the spiral wound it looks like that, that we have some sheets of the membrane and the sheets are separated by some this spacers and these sheets are then wound like a cylinder, and we find that on one side of the membrane we have the feed flow and between the membrane and the spacer, we have the permeate flow. So, here we have; that means, we have a bunch of these pairs of the membranes separated by the spacers, and that is how we are able to make a very compact module in the spiral form. So, this is the spiral wound configuration. Next we have the hollow fibre membrane module.

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In this case it is something like the shell and tube heat exchanger, in which the membrane comes in terms of the fibre or what we call lumen, and in this case we find that on the one side the feed enters then it goes through each of the fibres and then all the permeates are collected from one side, and on the other side we have the retentate which is going from the other side of the module. So, this is the overall configuration of the hollow fibre membrane module.

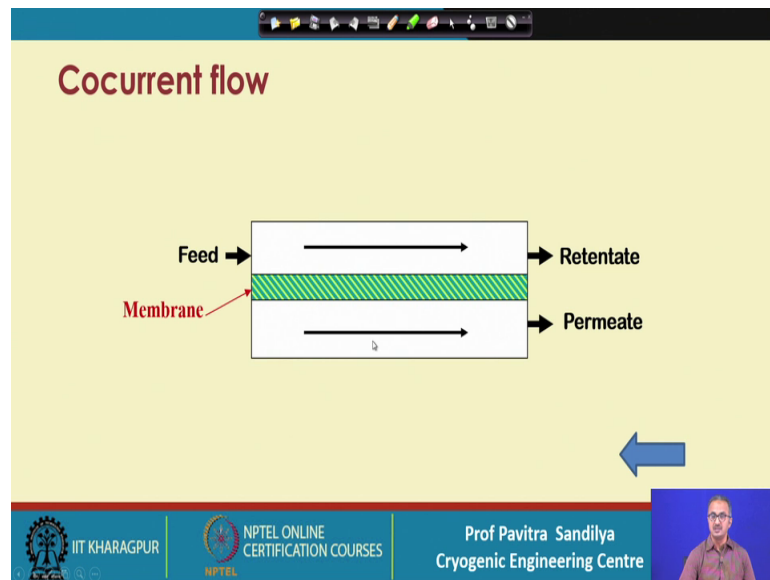
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The slide is titled "Flow patterns in membrane modules" in a dark red font. Below the title, there is a list of three flow patterns, each preceded by a checkmark and underlined in blue: "Cocurrent flow", "Countercurrent flow", and "Cross flow". The slide has a yellow background. At the bottom, there is a blue footer bar containing the IIT Kharagpur logo, the NPTEL logo, the text "NPTEL ONLINE CERTIFICATION COURSES", the name "Prof Pavitra Sandilya" and "Cryogenic Engineering Centre", and a small video inset of the professor.

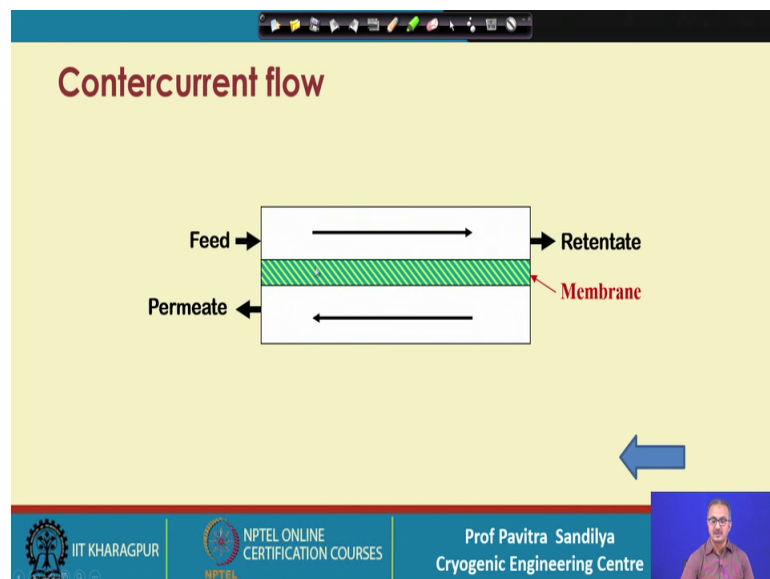
After the membrane modules, let us look in the various flow patterns possible in these modules; first you have the cocurrent flow, then you have countercurrent flow and we have the cross flow.

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So, go let us go one by one. In this cocurrent flow we find that feed enters at one place and it is decided based on the direction in which the permeate flow. So, in this case we find that feed and the permeate are moving on the same direction so that the retentate and permeate are taken out from the same side and this green represents the membrane.

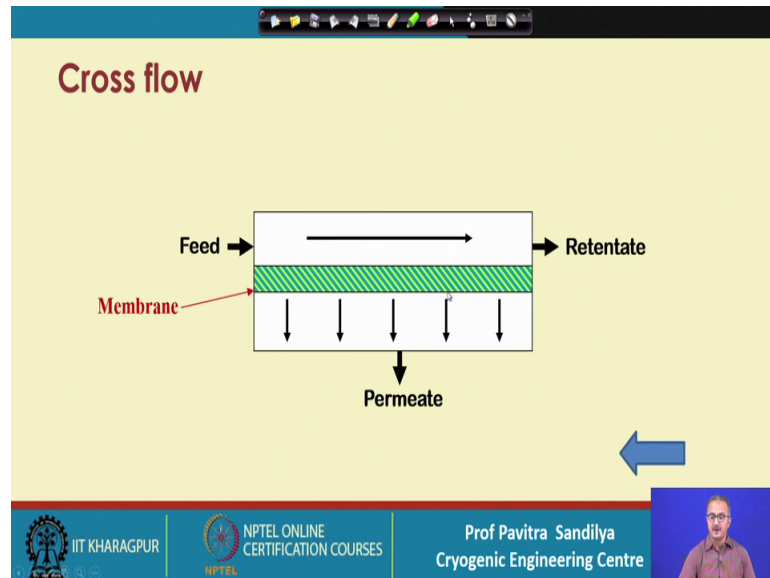
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Going to the countercurrent flow, we have this pattern that this retentate is taken from one side and the permeate is flowing countercurrent through that.

So, the to the opposite sides of the membrane, we are getting the retentate and the permeate whereas, in the cross flow.

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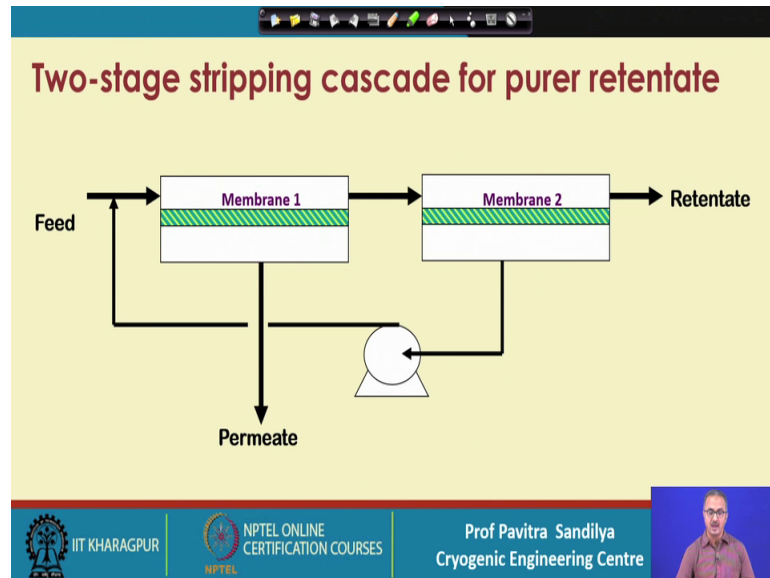
We find this kind of flow pattern that the feed it goes above the membrane and then the permeate is coming at a cross that is a 90 degree from the membrane. So, that is how we are getting a cross flow.

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The slide is titled "Membrane cascade" and contains a single bullet point: "✓ A single membrane module or a number of such modules arranged in parallel or in series without recycle constitutes a single-stage membrane separation process." The bottom of the slide features logos for IIT KHARAGPUR and NPTEL ONLINE CERTIFICATION COURSES, along with the name "Prof Pavitra Sandilya" and "Cryogenic Engineering Centre".

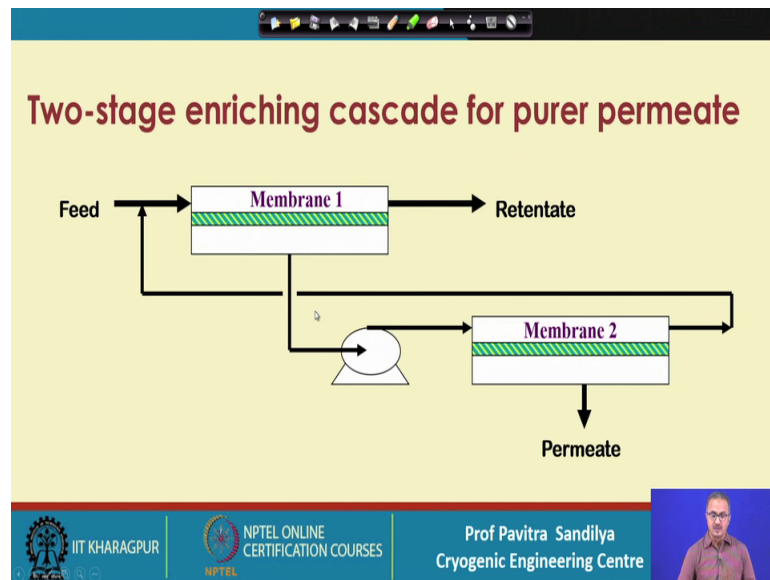
Now, we for a membrane cascade what we cascade means that, we are putting several membrane modules in series or parallel so that we can get a higher separation and in this case there could be a recycle in between the various modules.

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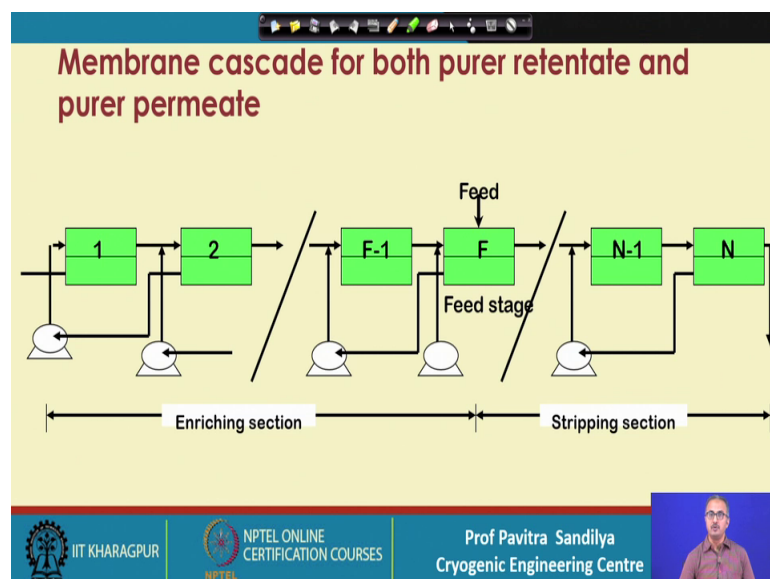
So, here we have one two stage stripping cascade, this is used for getting purer retentate. How? We find that the feed goes to the first membrane module here and the retentate is taken to the second module and then is getting the purer module; that means, this retentate and this retentate will be having different compositions whereas, the permeate is left free only thing may be that the permeate from the second model may be recycled to the first module 2 and take out some more retentate.

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Then we have a two-stage cascade for the purer component, in this case we find that what we are doing purer permeate. So, what we are doing that, we are now taking the permeate from the first module and taking it as a feed to a second module to get a purer permeate whereas, retentate is taken from the second module, and is fed back and mixed with the feed to send it back to the first module, and that is how we are trying to get a purer permeate.

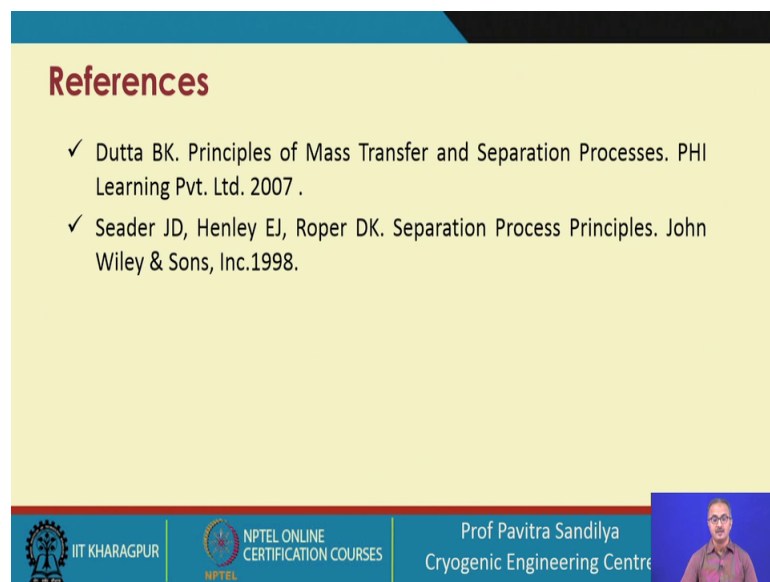
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And if we want both purer retentate and permeate like distillation column, we are what we are doing that, we are putting the feed somewhere in between and on the one side we are taking the retentate and putting it through the several modules and this we can say this is equivalent to the stripping section. And the on the other hand we take the permeate and this permeate is somewhat recycled and it is taken to the then the next module on this side, and this way we are finding that the permeate from the previous module is taken to the feed.

So, we find that this way we are able to get purer and purer permeate. So, this we can say this is something like an enriching section. So, this is how we are able to get a purer permeate and a purer retentate.

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References

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Now these are the references, which you may consider for further detail on the membrane.

Thank you.