



International Energy Agency IEA Implementing Agreement on District Heating and Cooling, including the integration of CHP

New materials and constructions for improving the quality and livetime of district heating pipes including joints - thermal, mechanical and environmental performance. New materials and constructions for improving the quality and lifetime of district heating pipes including joints – thermal, mechanical and environmental performance

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General Preface Annex VIII

Introduction

The International Energy Agency (IEA) was established in 1974 in order to strengthen the cooperation between member countries and reduce the dependency on oil and other fossil fuels. Thirty years later, the IEA again drew attention to serious concerns about energy security, investment, the environment and energy poverty. The global situation is resulting in soaring oil and gas prices, the increasing vulnerability of energy supply routes and ever-increasing emissions of climate-destabilising carbon dioxide.

At the 2005 Gleneagles G8 an important role was given to the IEA in advising on alternative energy scenarios and strategies aimed at a clean, clever and competitive energy future. Two years later, at the Heiligendamm G8, it was agreed that "instruments and measures will be adopted to significantly increase the share of combined heat and power (CHP) in the generation of electricity". District Heating and Cooling is an integral part of the successful growth of CHP: heat networks distribute what would otherwise be waste heat to serve local communities.

The IEA is active in promoting and developing knowledge of District Heating and Cooling: while the DHC programme (below) itself is the major global R&D programme, the IEA Secretariat has also initiated the International DHC/CHP Collaborative the kick-off event of which took place in March 2, 2007 with a 2-year Work Plan aiming to raise the profile of DHC/CHP among policymakers and industry. More information on the Collaborative is to be found on IEA's website www.IEA-org.

The major international R&D programme for DHC/CHP

DHC is an integrative technology that can make significant contributions to reducing emissions of carbon dioxide and air pollution and to increasing energy security.

The fundamental idea of DHC is simple but powerful: connect multiple thermal energy users through a piping network to environmentally optimum energy sources, such as combined heat and power (CHP), industrial waste heat and renewable energy sources such as biomass, geothermal and natural sources of heating and cooling.

The ability to assemble and connect thermal loads enables these environmentally optimum sources to be used in a cost-effective way, and also offers ongoing fuel flexibility. By integrating district cooling carbon-intensive electrically-based air-conditioning, rapidly growing in many countries, can be displaced.

As one of the IEA's 'Implementing Agreements', the District Heating & Cooling programme is the major international research programme for this technology. Active now for more than 25 years, the full name of this Implementing Agreement is 'District Heating and Cooling including the integration of Combined Heat and Power'. Participant countries undertake co-operative actions in energy research, development and demonstration.

Annex VIII

In May 2005 Annex VIII started, with the participation from Canada, Denmark, Finland, the Netherlands, Norway, South Korea, Sweden, United Kingdom, United States of America.

Below you will find the Annex VIII research projects undertaken by the Implementing Agreement "District Heating & Cooling including the Integration of Combined Heat and Power".

Project title	Company	
New Materials and Constructions for Improving the Quality and Lifetime of District Heating Pipes including Joints – Thermal, Mechanical and Environmental Performance	Chalmers University of Technology Project Leader: Ulf Jarfelt	8DHC-08-01
Improved Cogeneration and Heat Utilization in DH Networks	Helsinki University of Technology Project Leader: Carl-Johan Fogelholm	8DHC-08-02
District Heating Distribution in Areas with Low Heat Demand Density	ZW Energiteknik Project leader: Heimo Zinko	8DHC-08-03
Assessing the Actual Energy Efficiency of Building Scale Cooling Systems	International District Energy Association Project leader: Robert P. Thornton	8DHC-08-04
Cost Benefits and Long Term Behaviour of a new all Plastic Piping System	NUON Project leader: Hans Korsman	8DHC-08-05

Benefits of membership

Membership of this implementing agreement fosters sharing of knowledge and current best practice from many countries including those where:

- DHC is already a mature industry
- DHC is well established but refurbishment is a key issue
- DHC is not well established

Membership proves invaluable in enhancing the quality of support given under national programmes. Participant countries benefit through the active participation in the programme of their own consultants and research organisations. Each of the projects is supported by a team of

experts, one from each participant country. As well as the final research reports, other benefits include sharing knowledge and ideas and opportunities for further collaboration.

New member countries are very welcome - please simply contact us (see below) to discuss.

Information

General information about the IEA Programme District Heating and Cooling, including the integration of CHP can be obtained from our website <u>www.iea-dhc.org</u> or from:

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The IA DHC/CHP, Annex VIII, also known as the Implementing Agreement District Heating and Cooling, including the Integration of Combined Heat and Power, functions within a framework created by the International Energy Agency (IEA). Views, findings, and publications of the IA DHC/CHP do not necessarily represent the views or policies of all its individual member countries nor of the IEA Secretariat.

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Summary

PET foam has the potential to compete successfully with cyclopentane blown PUR foam as insulating foam for district heating pipes of small dimensions at low temperatures (<100°C).

Mechanical performance

The mechanical performance has been studied using the European standard EN 253:2003 as the reference. This standard is written with respect to PUR foams for pre-insulated district heating pipes. The standard applies for continuous operation at 120 °C for 30 years. However, for pipes of small dimensions, outer diameter \leq 120 mm, both temperatures and mechanical loads are lower.

- Compressive strength

The short-term compressive strength decreases with increasing temperature with a drastic change at approximately 80°C. This is due to the material's glass transition temperature. It can be seen that the compressive strength at high temperatures does not fulfil the requirements of 0.3 MPa for the PUR foam according to EN 253. It should be kept in mind, however, that this requirement is likely unnecessarily conservative for small pipes.

The short-term elastic modulus behaves in a similar fashion, with an obvious temperature dependence and a significant drop of stiffness around the glass transition temperature.

- Water permeability and vapour resistance

The tested PET foams were impermeable to liquid water and only vapour diffused through the PET foam. The vapour resistance was approximately 10 times greater for the PET foam than for a regular PUR foam.

- Water absorption

The water absorption for PET foam was found to be 7 %- vol. The requirement on PUR foam in EN 253 is a maximum of water absorption of 10 %. PUR foam usually absorbs around 5 %.

- Glass transition temperature

The results verify that the PET foams turn softer at approximately 80 °C, as could also be seen from the short-term compressive strength measurements. It is not possible to make a direct comparison with PUR. PUR is a thermoset material which does not undergo this kind of phase transition.

- Creep behaviour

The creep properties of PET foam seem to be very good. Extrapolation of the creep curves to 30 years of technical service does not indicate any significant creep deformation, neither at room temperature nor at 80 °C.

- Flexibility – bending properties

Samples of PET taken parallel to the extrusion direction are much stiffer and much more brittle, while samples taken parallel to the cross direction are so flexible that no fracture was seen up to the testing limit of 12 % strain. The material can easily be processed to withstand bending strains to a sufficient degree for flexible district heating pipes.

The PUR foams behave in a little more brittle manner. The "flexible" PUR foam did not exhibit a significantly more flexible behaviour than the rigid PUR foam. A strain limit of approximately 11 % was seen for both materials. It is also interesting to note that the flexible foam is much stiffer than the rigid foam.

Insulating performance

A newly produced PUR foam has a little lower thermal conductivity than a new PET foam (both cyclopentane blown and of the same density). The difference depends mainly upon smaller cells in the PUR foam and a lower content of air in the cell gas. In microcellular PUR foams the cell sizes are around 0.1-0.2 mm and in the PET-foams studied the cell size varies between 0.6 to 1.0 mm. However, the PUR foam has been developed during more than 50 years but the PET foam during less than 10 years and can thus be expected to be further developed. In the future a new PET foam will probably exhibit the same thermal conductivity as a PUR foam.

The results from the determination of the diffusion properties give a clear cut indication that the long term thermal performance of a PET foam is better then that of a PUR foam. The effective diffusion coefficients of oxygen, nitrogen and carbon dioxide in a PET foam are about 5-15 times lower than those in a PUR foam.

The long term thermal conductivity has been calculated for three different district heating pipes of the same dimensions (DN 40/125): 1. PUR foam insulation, 3 mm thick polyethylene casing, 2. PET foam insulation, 1 mm thick PET casing and 3. PET foam insulation without any casing. Due to the slow diffusion in the PET foam, the decrease of insulating capacity of the PET foam insulated pipe without casing is even less (about 6%) than that of the PUR insulated pipe with 3 mm HDPE casing (about 16%). The PET insulated pipe with 1 mm PET casing exhibits the slowest decrease (about 3%) of insulating capacity during 30 years among the three alternatives studied.

Environmental performance

From the present study it can be concluded that PET foam has the potential to compete successfully in terms of environmental performance with cyclopentane blown PUR foam as insulating foam for district heating pipes. The possibility to produce cyclopentane blown PET foam of low density will increase the competitiveness of PET foam. Unfortunately this possibility did not exist at the time of the environmental study. Commercial methods to produce PET foam for the insulation of district heating pipes must be developed. Utilization of recycled PET can reduce the environmental impacts from the production phase of the pipes life cycle and would contribute to the efficient use of resources in society.

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1. Introduction

1.1 Objectives / goals

New materials and constructions for district heating pipes must be very efficient from different points of view: Economical, thermal, mechanical and environmental.

In this project the efficiency of polyethylene terephthalate (PET) foam has been studied. According to the aspects mentioned above PET foam seems to have a great potential as a possible replacement alternative to polyurethane (PUR) foam in district heating pipes:

- Economy
 - The prices of PET and PUR foam have been about the same during the last years.
 - The possibility of utilising recycled material in the production of PET foam will decrease material costs.
- Thermal (Insulating) performance
 - If a PET and a PUR foam can be produced with the same blowing agent and have the same cell size and density, the thermal conductivity will be about the same. However, since the insulating foam will be used for a very long time, the foam exhibiting the slowest gas diffusion will have the best long term thermal performance (LTTP).
- Mechanical properties
 - Adequate mechanical properties are needed for a long lifetime.
- Environment
 - Superior insulation capacity during a long time is important in order to minimize the heat losses and the environmental impact (e.g. carbon dioxide emissions) caused by the energy production to compensate for heat losses.
 - The use of recycled material in the production of PET foam will increase resource efficiency.
 - There is a need to find alternatives to PUR foam, due to the toxicity of the isocyanates, one of the main components in PUR foam production. Another aspect is that welding close to PUR foam may give rise to high concentrations of hazardous compounds in the work environment.

1.2 Earlier work

PET has been used for a long time for textile fibres, bottles and film but foam made of PET is a fairly new material, still under development. Thin boards of PET foam have been used for e.g. packaging, especially in food industry, and for insulation applications. However, it has not been possible to produce thick boards of PET foam earlier. Today the Italian company B.C. FOAM has developed a technique for production of foam boards up to 90-95 mm thick.

An introductory study on the mechanical properties of PET foam revealed a higher compressive strength than required for PUR foam. Measurements of thermal conductivity showed that low density PET foams with a large fraction of blowing agent in the cells have the potential to reach the same low thermal conductivity as that of PUR foams [Mangs, 2005].

1.3 Properties of polyethylene terephthalate

Polyethylene terephthalate (PET) is a <u>thermoplastic polymer</u> resin of the <u>polyester</u> family and is used in <u>synthetic fibres</u>; beverage, food and other liquid containers; thermoforming applications; and engineering resins often in combination with glass fiber. It is one of the most important raw materials used in man-made <u>fibres</u>.

Depending on its processing and thermal history, it may exist both as an <u>amorphous</u> (transparent) and as a semi-<u>crystalline</u> (opaque and white) material. In discussing textile applications, PET is generally referred to as simply "polyester" while "PET" is used most often to refer to packaging applications.

It is manufactured under trade names e.g.: <u>Hostaphan</u>, <u>Melinex</u> and <u>Mylar</u> films, and <u>Dacron</u>, <u>Diolen</u>, <u>Terylene</u> and Trevira fibres.

Modulus of elasticity Tensile strength	2800 – 3100 MPa 55 – 75 MPa
Tensile strength	55 – 75 MPa
	55 75 Mil u
Glass temperature	75 °C
Melting point	260 °C
Thermal conductivity	$0.24 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$
Linear expansion coefficient	$7 \cdot 10^{-5} \text{ K}^{-1}$
Specific heat	$1.0 \text{ kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$

1.4 District heating pipe production

PUR foam insulated pipes

Discontinuous and continuous production of PUR foam insulated district heating pipes exists. In the discontinuous method each pipe is manufactured individually in lengths of 6 to 16 m. The thickness of the PUR foam for each dimension can be varied according to the intended application. The high density polyethylene (HDPE) casing is first produced in a continuous process where HDPE granules are melted and extruded into a cylindrical shape and then cut to the desired length. The media pipe is centred in the casing, after which the PUR foam formulation is injected through a hole in the casing in the middle of the pipe or from one of the ends. The foam is created as a result of chemical reactions of the components in the formulation. When the PUR foam expands, it adheres to the surface of the steel pipe and the HDPE casing.

There are two types of continuous production techniques. In the "spray process", layers of PUR foam formulation are sprayed onto the steel pipe. The HDPE casing is either extruded or wound around the insulation. In the continuous moulding process, the foam formulation is laid on a polyethylene sheet with the steel pipe centred above it. The sheet is pulled into a pipe-shaped mould as the PUR foam expands. The HDPE casing is then extruded onto the pipe. Finally the pipes are cut to the desired lengths.

PET foam insulated pipes

Production of PET foam insulated district heating pipes is still being developed. Boards of a thickness up to about 90-95 mm can be produced in an extrusion process developed by the Italian company B.C. Foam. So far PET foam for pipe insulation has only been produced for small dimensions and in a small scale.

1.5 Polyurethane

Polyurethane is a widely used material. In Western Europe in 2003, PUR constituted about 5% or 2.7 million tons of the total annual consumption of plastics. There has been a steady increase since 1995, when consumption was 1.8 million tons [APME 1997, 2000, 2001, 2002, 2003, 2004]. The great possibilities of adjusting the material properties, e.g. density, flexibility and stiffness, opens up for a variety of different applications, such as coatings in the appliance industry, soles for shoes, glues, mattresses, vehicle seats and insulation. In many countries rigid PUR foam is commonly used as insulation material in the construction industry due to its low thermal conductivity and the fact to that it can be foamed on site. PUR foam is formed by simultaneous polymerisation and expansion in a formulation containing an isocyanate, a polyol and a blowing agent with a low boiling point (Figure 1). The polyol contains two or more hydroxyl groups.

Figure 1. Polymerisation reaction of polyurethane



For thermal insulation applications, the heat conduction in the solid polymer must be minimized, thus the density should be kept as low as possible without jeopardising the strength of the foam. Normally, the density of the PUR foam in district heating pipes is about $60-70 \text{ kg} \cdot \text{m}^{-3}$.

Figure 2. Demand (= all virgin polymers used by converters in Europe including imports) of different plastics in Europe in 2005 [Plastics Europe, 2007]. "PE-(L)LD(HD)" =(linear) low(high) density polyethylene, "PP" = polypropylene, "PS" = polystyrene (expandable grades not included), "PVC" = polyvinyl chloride, "PET" = polyethylene terephthalate (bulk), "PUR" = polyurethane (for 2003, from APME 2004).



1.6 Polyethylene terephthalate

There has been a worldwide increase in the consumption of PET during the last decade [APME 2004, 2005]. Between 1995 and 2001 the total consumption of PET in the world increased from about 3 million to almost 8 million tons. PET used for bottles and film in Europe has increased almost threefold since 1995, while the use of the material for textile fibres has remained more or less static (Figure 3).



PET recycling is also expected to increase due to the European Commission's Packaging and Packaging Waste Directive, which includes higher targets for mechanical recycling of plastic packages all over Europe [European Council 1994 and 2005]. PET is a thermoplastic and can thus be remelted into new products. Since production of food packages from recycled material is restricted for hygienic reasons, new fields of application for recycled PET such as foam production, can be developed.

PET (Figure 4) was developed by the British Calico Printers company 1941 and was originally used for synthetic fibres [Bousted 2002]. Films have been produced since the mid-1960s and the PET bottle blowing technique was introduced in the 1970s.



Figure 4. Polyethylene terephthalate



Polyesters are obtained by a reaction between an acid and an alcohol (Figure 5). Terephthalic acid or dimethylterephthalate and ethylene glycol are used in the production of PET and the reaction takes place in the presence of a catalyst. *p*-Xylene derived from catalytic reforming of naphta is used to produce terephthalic acid and dimethylterephthalate. Ethylene made from cracked natural gas or the naphta fraction from crude oil is used to produce ethylene glycol [Bousted 2002]. For both reaction routes a polycondensation reaction follows. A short liquid state polycondensation creates an amorphous material suitable for fibres and film. Semi-crystalline material for bottles and PET foam is achieved after a longer period of polycondensation [Xhanthos 2000].





The production of PET foam boards is shown in Figure 6. The polymer granules are dried and mixed with a nucleating agent (talcum 0.5 % by mass) before entering the extruder. The blowing agent (approx. 1 % by mass) is mixed under high pressure (70-80 bar) into the polymer melt in the extruder and kept under pressure until the melt exits through the die where the extrusion pressure is about 45-50 bar. In the atmospheric pressure following the die lips, the blowing agent is transformed from the liquid to the gaseous phase, creating the foam cells.

The price of virgin materials follows the fluctuations in the cost of raw oil. During the last years the prices of PET and PUR have been about the same (2.03 EUR/kg [PET] and 1.95-2.0 EUR/kg [PUR], August 2007). The price for recycled PET of good quality was significantly lower, around 0.8 EUR/kg. Recycled PET has, however, been shown to be unsuitable for foam production, due to contamination of other plastics and degradation of the material during the recycling process [Japon 2000, Xhanthos 2000]. The problem is that the viscosity of recycled PET is too low and the melt strength inferior compared to virgin material. Foams of a very high quality have been achieved in experiments where recycled PET was chemically modified by means of multifunctional branching agents (e.g. tetraglycidyl diamino diphenyl methane or pentaerythritol) [Japon 2000, Smith 1990, Xhanthos 2000].

Figure 6. Schematic illustration of a process plant used to produce PET foam sheets [M&G Polymers 2004]



Studies show that PET undergoes chemical degradation by hydrolysis if exposed to humidity or water at elevated temperatures which results in increased embrittlement [Foulc 2005, Oreski 2005]. In district heating pipes, this could become a problem for the PET foam close to the media pipe where the temperature is elevated. Thus, PET foam may preferably be used for district heating pipes of small dimensions where the temperature normally is lower.

One function of the HDPE casing in district heating pipes is to protect the PUR foam insulation from external mechanical pressure. Due to the high compressive strength (see 2.2.3) of PET foam it may be possible to use PET foam insulated pipes without a casing or with a casing of PET. Production techniques have not yet been developed, but the casing could, for example, be produced by melting the outer layer of the insulation. Fewer materials in a product make the waste treatment of pipes taken out of use considerably easier.

1.7 Blowing agents

Table 1 presents some technical and environmental features of previously and currently used blowing agents in PUR and PET foam insulations. It is desirable that a blowing agent has a fairly high vapour pressure, thus making it possible to introduce more gas in the foam. A high gas content leads to a slow deterioration of the insulation capacity over time. The thermal conductivity of the blowing agent has a major impact on the foam thermal conductivity and should be as low as possible to ensure foam with good insulating capacity.

The atmospheric lifetime is an important environmental factor, since substances with a long life can potentially cause more environmental damage such as ozone depletion and global warming. The ozone depletion potential (ODP) for the blowing agents with CFC-11 as a reference substance and the global warming potential (GWP) with carbon dioxide as reference substance are presented in Table 1.

The chlorofluorocarbons (CFCs) and carbon dioxide were used in the first types of PUR foam insulated district heating pipes. The effect of the CFCs on the ozone layer led to a shift towards hydrochlorofluorocarbons (HCFCs) and carbon dioxide. In a PUR foam carbon dioxide will always be present since it is formed in the reaction between isocyanate and traces of water always present. Today, cyclopentane in combination with carbon dioxide, is the main blowing agent in European PUR foam insulated district heating pipes. In the United States pentafluoropropane (HFC-245fa) is used as replacement for HCFCs. Pentafluorobutane (HFC-365mfc) is a gas that has been considered for European district heating pipes.

In the development of PET foam, tests with different blowing agents have been performed in pilot plants. The gases used have been carbon dioxide, difluoroethane (HFC-152a), chlorodifluoroethane (HCFC-142b) and chlorodifluoromethane (HCFC-22).

The main disadvantages of carbon dioxide is its high thermal conductivity (16.3-16.6 mW·m⁻¹·K⁻¹ at 25 °C) compared to other blowing agents (Table 1) and its fast diffusion out of the foam leading to a rapid decrease in insulation capacity. Solely carbon dioxide blown PUR foam is normally only used for pipes of large dimensions. Cyclopentane has lower thermal conductivity than carbon dioxide (11.3-13.8 mW·m⁻¹·K⁻¹ at 25 °C) as well as a much slower rate of diffusion. The vapour pressure of cyclopentane is below 1 bar at room temperature (0.42 bar at 25 °C) [Chemnetbase 2005]. In addition to cyclopentane,

isopentane with a vapour pressure of 0.917 bar at 25 °C and a thermal conductivity of 14.8 $mW \cdot m^{-1} \cdot K^{-1}$ at 25 °C [Chemnetbase 2005, Volkert 1995] is sometimes added into the foam formulation in order to increase the total amount of hydrocarbons in the foam cells. A disadvantage of hydrocarbons is their flammability and thus extra precautions must be taken during handling and processing operations. Although produced from fossil resources, hydrocarbons have no ozone depletion effect and very low global warming effect. A report by Galvin et al. summarizes the results of toxicological studied carried out on rats, mice and rabbits since the 1940s [Galvin 1999]. From these studies it was concluded that cyclopentane is practically non-toxic and has no significant mutagen effects.

Hydrofluorocarbons (HFCs) have been used as replacement options in several technical applications including PUR foam insulation. The gases in this group are non-ozone depleting but have rather high global warming potentials, up to 10,000 times higher than carbon dioxide [IPCC 2001a, Naik 2000].

In the USA, HCFCs used for production of new insulation foams were phased out in 2003, and HFC-245fa (1,1,1,3,3-pentafluoropropane, Enovate TM3000) is produced and marketed as a replacement option by the American company Honeywell. As it is a non-flammable gas, it is possible to use the same equipment as for the production of PUR foam blown with dichlorofluoroethane (HCFC-141b) [Bogdan 2001]. The German company Bayer AG has patents in Europe, the USA and Canada for methods to produce PUR foam with HFCs as blowing agent [Bayer AG Leverkusen 1990, 1996]. Honeywell has acquired a world wide licence from Bayer AG with exclusive rights for USA and Canada, while Solvay has acquired a world licence excluding the USA and Canada. Solvay produces a range of

HFCs marketed under the name Solkane®, of which HFC-365mfc is one of their main products intended for PUR foam insulation [Solvay Fluor 2005].

Due to the flammability of HFC-365mfc, the Solvay company has developed non flammable mixtures with 7 % and 13 % heptafluoropropane (HFC-227ea). At Solvay's French production plant in Tavaux, all hydroflouroalkanes are manufactured by means of hydrofluorination of a chlorinated precursor [Zipfel 1999]. At the end of 2002, a plant with an annual HFC-365mfc production capacity of 15,000 tons commenced operation in Tavaux in France. The price of HFC-365mfc and HFC-245fa is higher than that of cyclopentane. Since the chemicals used for the PUR formulation differ due to the blowing agent used, the total cost must be considered.

The vapour pressure of HFC-365mfc and HFC-245ea is higher than that of cyclopentane, which may result in a higher concentration of these gases in the foam. The thermal conductivity of HFC-365mfc (10.6 mW·m⁻¹·K⁻¹ at 25 °C) is of about the same as that of cyclopentane and lower than that of HFC-245fa (12.2-13.5 mW·m⁻¹·K⁻¹ at 25 °C) (Table 1). When HFC-365mfc is mixed with HFC-227ea, the thermal conductivity of the gas mixture increases to 10.7-10.9 mW·m⁻¹·K⁻¹ at 25 °C [Zipfel 2002].

A toxicological study of HFC-365mfc showes low toxicity and no significant mutagenicity [Zipfel 1999]. Studies on HFC-245ea show that the substance is of low toxicity, non-mutagenic and not a teratogen [Honeywell 2005].

	Carbon dioxide	Cvclopentane	HFC-365mfc	HFC-245fa	HFC-152a	HCFC-142b	HCFC-22	CFC-11
		\bigcirc	F F F F	F F F F F	F	F F Cl	F F CI	
	<i>CO</i> ₂	C_5H_{10}	CF ₃ CH ₂ CF ₂ CH ₃	CHF ₂ CH ₂ CF ₃	CHF ₂ CH ₃	CClF ₂ CH ₃	CHClF ₂	CCl ₃ F
CAS registry no	124-38-9	287-92-3	406-58-6	460-73-1	75-37-6	75-68-3	75-45-6	75-69-4
Molecular weight (g·mole ⁻¹)	44.0	70.1	148.1	134.0	66.1	100.5	86.5	137.4
Boiling point (°C)	-78.5 Ref: a	49.3 Ref: a	40.2 <i>Ref: b</i>	15.3 <i>Ref: r</i>	-25 to -24.1 Ref: a,c,d,e,l	-10 to -9.2 <i>Ref: a,c,d,f</i>	-40.8 Ref: a,c,d	23.7 <i>Ref: a</i>
Vapour pressure @ 25 °C (bar)	64.3 Ref: c	0.42 Ref: e	0.47 (20°C) Ref: b	1.23 (20°C) Ref: r	6.0-6.1 Ref: c,d,e	3.4-3.5 <i>Ref: c,e</i>	10.4 Ref: c,e	1.1 <i>Ref: e</i>
Thermal conductivity of gas phase @ 25 °C (mW·m ⁻¹ ·K ⁻¹)	16.3-16.6 <i>Ref: c,f,g,h</i>	11.3-13.8 Ref: i,h,v	10.6-11.6 <i>Ref: b,v</i>	12.2-14.8 <i>Ref: b,j</i>	13.4-14.7 Ref: c,d	9.4-12.1 Ref: c,f,j	10.6-11.7 Ref: c,f,h,j	7.4-8.2 Ref: f,h,j
Flammability limits in air (vol%)	None	1.5 – 8.7 <i>Ref: k</i>	3.5-13.3 <i>Ref: l,t</i>	None Ref: b	3.7-20.2 Ref: c,m,n	9.0-14.8 Ref: c,m,n	None Ref: c	None Ref: f
Atmospheric lifetime (years)		< 0.01 Ref: u,s	8.6-10.7 Ref: o,p,q	7.2-8.4 <i>Ref: o,p,q,s</i>	1.4-1.8 Ref: n,o,p,q	17.9-22.4 <i>Ref: n,o,p,q</i>	11.9-15.8 Ref: n,o,p,q	45-50 Ref: o,p,q
Ozone Depletion Potential, 100 years	0	0	0	0	0	0.014-0.07 <i>Ref: q</i>	0.034-0.055 <i>Ref: q</i>	1
Global Warming Potential, 100 years	1	< 10 Ref: u	782-953 Ref: o,p,q	950-1020 Ref: o,p,q	120-149 Ref: o,p,q	1957-2400 Ref: o,p,q	1700-1780 Ref: o,p,q	4600-4749 Ref: o,p,q
 a) [Knovel Corp 2005] b) [Zipfel 2002] c) [Albouy 1998] d) [Creazzo 1995] e) [Chemnetbase 2005] f) [Shankland 1990a] 		 g) [L'air liquide h) [Brodt 1995] i) [Volkert 1995] j) [Heinemann k) [Galvin 1999] l) [Solvay Fluo 	2 1976]] 5] 2000] 9] r 2005]	 m) [Decaire 19 n) [Barthélemy o) [Naik 2000] p) [IPCC 20011 q) [World Meter r) [Honeywell 2 	94] 1993] 5] corological Org 2 2005]	002]	s) [Seifert 2003] t) [Zipfel 1999] u) [Heilig 1994] v) [Merten 1997]	

Table 1. Physical and environmental properties of different insulating gases used as blowing agents in PET and PUR insulation foam

2. Methods and results

2.1 Development of the PET foam

Before the start of this project many different blowing agents were used for the PET foam production. HCFC-142b, HCFC-22, HFC-152a, carbon dioxide and nitrogen were used due to their satisfactory performance in the foaming process. The lowest density of foam board produced at that time, about 85 kg·m⁻³, was achieved with a mixture of HCFC-142b and HCFC-22 [Mangs 2004]. However, these gases cannot be considered as a solution for the future due to their ozone depletion potentials.

HFC-152a, used for foams with densities in the range 95 - 120 kg·m⁻³, has about the same thermal conductivity as cyclopentane as well as a somewhat lower global warming potential than the other HFCs (Table 1). Inhalation experiments on rats indicate that HFC-152a has very low acute toxicity [Keller 1996].

Carbon dioxide blown foam has been produced with densities down to about 140 kg·m⁻³.

Nitrogen has been used for PET foam up to densities of 400 kg·m⁻³. These foams are not intended to be used for insulation purposes, but as construction material.

An important breakthrough within this project happened when cyclopentane could be used as a blowing agent. B.C. Foam succeeded in producing thick foam boards (about 60 mm, see Figure 7) with densities down to $55 \text{ kg} \cdot \text{m}^{-3}$. It was also possible to manufacture limited quantities of a pipe foam jacket of small dimensions (outer diameter: 75 mm, inner diameter 18 mm, see Figure 8).



Figure 7. Foam board of PET produced by B.C. Foam. Thickness about 65 mm

Figure 8. Pipe PET foam jacket produced by B.C. Foam. Blowing agent: cyclopentane. Foam density: 55 kg·m⁻³. Outer diameter: 75 mm, inner diameter: 18 mm.



2.2 Mechanical performance

Tests have been performed at room temperature and at elevated temperatures on PET materials stored at room temperature. As far as possible, the test scheme according to the European Standard 253, EN 253:2003, has been followed. All mechanical tests were performed at the Swedish Research Institute.

The foam boards were manufactured by B.C. Foam. The foams were designated A - D. The following blowing agents were used:

Material	Blowing agent
А	HFC-152a
В	HCFC-142b/HCFC 22
С	HFC-152a
D	Cyclopentane

The following material properties have been determined:

- 2.2.1 Density
- 2.2.2 Cell size
- 2.2.3 Compressive strength
- 2.2.4 Tensile strength
- 2.2.5 Water permeability and vapour resistance
- 2.2.6 Water absorption
- 2.2.7 *Glass transition temperature*
- 2.2.8 Creep behaviour
- 2.2.9 *Flexibility bending properties*

2.2.1 Foam density

The foam density of the materials was determined in accordance with EN 253:2003, clause 5.3.3, on specimens with dimensions 30 mm x 30 mm x 20 mm. The results, given in the table below, are mean values from measurements on at least three specimens of each material:

Material	Blowing agent	Density	
		kg·m⁻³	
А	HFC 152a	114	
В	HCFC 142b/HCFC 22	84	
С	HFC 152a	95	
D	Cyclopentane	69	

2.2.2 Cell size

The average cell size was determined in accordance with clause 5.3.2.1 of EN 253:2003. The cell size was analysed in the three principal directions from the board extrusion process, Figure 9.

Figure 9. Directions in analysed sample



The average size of the cells in the x, y and z direction is estimated by counting the sum of cells on a 10 mm long distance and then divide the distance with the sum of cells. The counting of the cells were accomplished by pressing the planes X, Y and Z into sheets of clay and taking photographs at close range. Figure 10 shows casts of the cells from the four materials. Table 2 shows the average cell sizes in the three directions of the materials.



Figure 10. Casts of material A (upper left), material B (upper right), material C (bottom left) and material D (bottom right). The squares at the bottom sections measure $1 \times 1 \text{ mm}^2$

Material Density (kg⋅m ⁻³)	Average cell diameter (mm) Direction		
	х	у	Z
A (114)	0.53	0.63	0.56
B (84)	0.52	0.61	0.65
C (95)	0.63	0.68	0.65
D (69)	0.70	0.88	0.91

Table2. Average cell sizes of the PET foams

The most prominent anisotropy is found in material D, which is slightly "flattened" in the cross direction X. The other materials did not exhibit any significant anisotropy.

2.2.3 Compressive strength

The compressive strength was analyzed for materials A, B, C and D on square samples with side length 30 mm and thickness 20 mm. Tests were made at the temperatures: 21, 35, 50, 60, 70, 80, 98, 120 and 140°C. The compression rate was 10 % per minute.



The results of the short term compressive strength tests are shown versus temperature as a diagram in Figure 12. From the diagrams it can be seen that the short-term compressive strength decreases with increasing temperature with a drastic change at approximately 80°C. This is due to the material's glass transition temperature, T_g . It can be seen that the compressive strength at high temperatures does not fulfil the requirements of 0.3 MPa on the PUR foam according to EN 253. It should be kept in mind, however, that this requirement is likely unnecessarily conservative for small pipes.



Figure 11. Sample of PET foam (a), sample in compression fixture (b), sample and fixture in climate chamber (c) and climate chamber in testing machine (d)

Figure 12. Short-term compressive strength vs. reciprocal temperature

The short-term elastic modulus behaves in a similar fashion, with an obvious temperature dependence and a significant drop in stiffness around the glass-transition temperature, Figure 13. It may also be noted that there seems to be a very linear correlation between the compressive strength and the short-term elastic modulus, Figure 14.





Figure 14. Short-term elastic modulus vs. short-time compressive strength

2.2.4 Tensile strength

The tensile strength was tested, at 20°C, on one test specimen from material D. The test was made on a tensile bar with a $16 \times 16 \text{ mm}^2$ square cross-section, Figure 15.



Figure 15. Test specimen for tensile strength

Brittle fracture occurred at 0.67 MPa. The strain at failure was 11 %. With the tensile curve, it is possible to establish the complete stress-strain relationship at room temperature, Figure 16.



Figure 16. Stress/strain curve for material D (69 kg·m⁻³) at room temperature

2.2.5 Water permeability and vapour resistance

The measurements were done on two test specimens from PET material A and on two test specimens from PET material B. The test specimens were 20 mm thick and shaped like disks with a diameter of 81 mm. The test specimens were installed in special cylinders. In the cylinder, one side of the specimen was exposed to water at 5 kPa overpressure. The other side of the sample was exposed to air at atmospheric pressure. A schematic view of a test cylinder is shown in Figure 17.

After applying the test specimen in the cylinder, the cylinder and all of its components were weighed. Then 50 ml water was put into the cylinder and then weighed again. Finally, after pressurizing the cylinder with 5 kPa, it was weighed a third time.



Figure17. Schematic view of test cylinder

The amount of water leaking or water vapour diffusing through the PET foam was determined by weighing the cylinders. The overpressure in the cylinder was monitored and adjusted periodically. During the test period the cylinders were stored at room temperature. At each weighing, the actual atmospheric pressure, actual room temperature and actual atmospheric humidity was measured.

The results showed that the tested PET foams from material A and B are impermeable to liquid water and only vapour diffused through the PET foam. The vapour resistance turned out to be approximately 10 times greater than for regular PUR foam, corresponding to a vapour diffusion coefficient of about $5 \cdot 10^8 \text{ m}^2 \cdot \text{s}^{-1}$. In Figure 18 the measured weight loss during the test period through the test specimens from the PET materials A and B is compared to the weight loss through regular PUR foam.



Figure 18. Measured weight loss due to vapour diffusion in 20 mm thick specimens taken from PET foam A and B compared to PUR foam.

2.2.6 Water absorption

Water absorption test was undertaken on three cube shaped samples with side lengths 25 mm from material D. The mass and volume of the specimens were determined. The cubes were then immersed in boiling water for 90 minutes, whereupon they were immersed in water at 23 °C for 60 minutes. Finally, after removal of surface water the final mass was determined and the water absorption calculated. Figure 19 shows the test cubes when boiling in water.

The water absorption for material D was established to 7.4 % (by volume). The requirement on PUR foam in EN 253 is a maximum of water absorption of 10 %, and regular PUR foam usually absorbs around 5 %.



Figure 19. Cubes of PET foam in boiling water

2.2.7 Glass transition temperature

The glass transition is a thermodynamic phase change. Above the glass transition temperature T_g , the inter-molecular bonds are weak and the material is soft and said to be in a rubbery state. Below T_g , the material is in a glassy state and acts in a stiffer manner. Analysis of the glass transition temperature was done according to the ASTM standard E 1356-98 for the PET materials A, B, C and D. The analyses were performed by differential scanning calorimetry with an apparatus of type Mettler Toledo DSC823^e, Figure 20.

Figure 20. Mettler Toledo DSC823^e



The method comprises the continuous monitoring of the difference in heat flow into a sample of the tested material and a reference sample when they are heated at a controlled rate through the glass transition region. By analysing the heat flow into the sample, the latent heat absorbed during the phase transition can be traced. Figure 21 shows the resulting thermal curves for the analysed PET materials. Table 3 shows the results from evaluating the curves according to a standardised procedure.

The results verify that the PET materials turns softer at approximately 80 °C, as could also be seen from the short-term compressive strength measurements in different temperatures, Figures 12 and 13.





It is not possible to make a direct comparison with PUR. PUR is a thermoset material and does not undergo this kind of phase transition. However, measurements on carbon dioxide blown PUR foam with a TMA (Thermo-Mechanical Analysis) method showed a softening temperature at around 110–120°C [Jarfelt et al. 1992].

Figure 21. Resultant thermal curves for the PET foams in the following order: C, A, D and B (from top to bottom)

Table 3. Glass transition region measured temperatures

2.2.8 Creep behaviour

The creep behaviour under compressive loading was measured on materials A and D at room temperature and at 80°C. The test specimens at room temperature were of dimensions $100 \times 100 \times 40 \text{ mm}^3$, and the dimensions of the specimens tested at higher temperature were $30 \times 30 \times 30 \text{ mm}^3$, Figure 22.



Figure 22. Creep testing at room temperature (left) and in climate chamber at $80 \, \text{°C}$ (right).

The creep samples were subjected to a compressive load corresponding to 30 % or 50 % of the short-term compressive strength (Figure 12) at the temperature in question.

The results are shown in Figures 23 and 24. The high temperature curves have been adjusted with respect to a zero-load reference. At elevated temperature, the increase in cell gas pressure will counteract the compression load. By measuring on a reference sample without load, a creep expansion can be seen. The high temperature curve for material D indicates a slight expansion also under loading after approximately 300 hours. The reason for this is not clear, but it is likely caused by cell gas effects.

However, it may be concluded that the creep properties of PET seem to be very good. Extrapolation of the creep curves to 30 years of technical service does not indicate any significant creep deformation. Furthermore, the creep rate at high temperature is well in line with PUR.
Figure 23. Creep deformation of material A and D at room temperature under compressive stress corresponding to 30 % and 50 % respectively of the short-term strength.



Figure 24. Creep deformation of material A and D at 80 $^{\circ}$ under compressive stress corresponding to 50 % of the short-term strength. Comparison with regular PUR foam at 100 $^{\circ}$ C.



2.2.9 Flexibility – bending properties

One of the most important benefits with flexible pipes is that they can be coiled and thereby manufactured in very long lengths. This allows for a joint-free construction and a rational laying. Consequently, an important property for a foam for flexible pipes is that it can withstand the bending strains induced by the coiling. In reality, a flexible pipe will be subject to repeated bending by coiling and un-coiling several times. However, the aspect of repeated bending has not been covered in the present study.

The bending strains due to coiling are illustrated in Figure 25. The outside circumference of the coil will be longer than the mean circumference, and this will lead to tensile strains. Similarly, compressive strains will arise along the inside circumference.



The bending strain (ϵ) level will increase with pipe diameter *d* and decrease with the coil's radius *R*:

$$\varepsilon = \frac{(R+d/2)\Delta v - R\Delta v}{R\Delta v} = \frac{d}{2R}$$

As an example, Logstor sells PEX flex pipes with a diameter of 77 mm in coils with minimum radius of 0.8 m, and 160 mm pipes in 1.0 m coils. This corresponds to approximately 5 % and 8 % bending strain respectively.

25

Figure 26. Bending tests on foam bars of length 200 mm and cross-section $\sim 15 \times 15 \text{ mm}^2$



The flexural properties have been evaluated by three-point bending tests at room temperature, largely in accordance with EN ISO 178, see Figure 26. Bar samples with a $15 \times 15 \text{ mm}^2$ square cross-section and a 200 mm span were used. A point-load was applied at the mid-span so that a flexural strain rate of 1 % per minute was achieved.

Tests were undertaken on the PET material D. This particular material has been manufactured as a board in an extrusion process and it is expected to be anisotropic, i.e., behave differently in different directions. In the principal board directions, there are four possible ways of extracting and loading bending samples, see Figure 27. For comparison, PUR foam samples taken from both a rigid straight pipe and a coiled flexible pipe were tested.



Sample designation y_x denotes bending around the y axis of a bar parallel to the x axis, etc. x is the direction of extrusion, y is the cross direction and z is the thickness direction.

The results from the bending tests are given as curves of bending stress vs. bending strain in Figure 28. There is clearly an influence from the direction. Samples taken parallel to the extrusion direction are much stiffer and much more brittle, while samples taken parallel to the cross direction are so flexible that no fracture was seen up to the testing limit of 12 % strain. The material can easily be processed to withstand bending strains to a sufficient degree.

Figure 27. The four sample orientations for bending tests on bars taken from a board configuration





The PUR foams behave in a little more brittle manner, see Figure 29. The "flexible" foam did not exhibit a significantly more flexible behaviour than the rigid foam. A strain limit of approximately 11 % was seen for both materials. It is also interesting to note that the flexible foam is much stiffer, than the rigid foam. The stiffness is the slope of the curve and it can be taken as a measure on how much force must be applied to coil or un-coil the pipe.

Sample bars were taken in the axial direction of a district heating pipe and loaded parallel to the cross-section radius. Flexible samples were taken from a coil at the crown of the cross-section. Hence, the tests were undertaken in a previously "un-bent" direction



Figure 29. Bending stress vs. bending strain on PUR foams from straight pipes and flexible pipes.

2.3 Insulating performance

2.3.1 Thermal conductivity

The thermal conductivity of a cellular foam can be described as the sum of the three main mechanisms, conduction in the cell gas mixture, conduction in the polymer matrix and radiation. The cell gas conduction is time (t) dependent due to the change of cell gas composition as a result of diffusion. All mechanisms are temperature dependent, but the dependency is more pronounced for cell gas conduction and radiation.

$$\lambda_{foam}(t) = \lambda_{gas}(t) + \lambda_{pol} + \lambda_{rad}$$

where	λ_{foam}	total thermal conductivity of the foam
	λ_{gas}	thermal conductivity due to gas conduction
	λ_{pol}	thermal conductivity due to conduction in the polymer matrix
	λ_{rad}	thermal conductivity due to radiation

Convection only occurs if the cell size is large (exceeds 5 mm) [Isberg 1988]. Convection is normally negligibly in the PUR foam used for district heating pipes and in the PET foam qualities studied.

Gas Conduction

The thermal conductivity due to conduction in the cell gas mixture can been calculated using Wassiljewa's equation with the Mason and Saxena modification [Mason 1958].



Symbols:

$W \cdot m^{-1} \cdot K^{-1}$
$W \cdot m^{-1} \cdot K^{-1}$
-
kg·kmole ⁻¹

The thermal conductivities of the pure gases used when calculating the conductivity of the gas mixture are presented in Table 4.

Table 4. Thermal conductivity of air, carbon dioxide and cyclopentane at 10°C and 50°C.

Gas	$\stackrel{\lambda_{10}}{W\cdot m^{\text{-1}}\cdot K^{\text{-1}}}$	$\stackrel{\lambda_{50}}{W\cdot m^{-1}\cdot K^{-1}}$	Molecular weight g·mole ⁻¹
Air ^[a] Carbon dioxide ^[a]	0.0250	0.0282	30
Cyclopentane ^[b]	0.0127	0.0155	70

^[a] Dognin, AS., Académie des Sciences, p 243, 1956.

^[b] Calculated from the value given at 25°C by Brodt, K., Thermal insulations: cfc-alternatives and vacuum insulation, Doctoral thesis, Technische Universiteit, Delft, 1995.

Conductivity in the polymer matrix

The contribution to the heat flow due to conduction in the solid polymer matrix is a combination of the heat flow in the cell walls and in the struts.

From experiments with crushed foam the thermal conductivity of the solid PUR polymer (λ_{solid}) was found to be 0.22-0.26 W·m⁻¹·K⁻¹ [Glicksman 1994]. For solid PET polymer a thermal conductivity of 0.24 W·m⁻¹·K⁻¹ has been reported (see 1.3).

The main part of the solid polymer is found in the struts. The fraction of solids in the struts (f_s) has been reported to 0.8 [Sinofsky 1984].

The thermal conductivity due to conduction in the polymer matrix has been described by [Sinofsky 1984].

$$\lambda_{pol} = \lambda_{solid} \cdot \frac{1}{3} \cdot f_s \cdot (1 - \delta) + \lambda_{solid} \cdot \frac{2}{3} \cdot (1 - f_s) \cdot (1 - \delta) \qquad \text{where} \quad \delta = 1 - \frac{\rho_f}{\rho_s}$$

Symbols:

f_s	fraction of solid in struts	-
δ	volume fraction of voids or cell interiors	-
λ_{solid}	thermal conductivity of the solid polymer	$W \cdot m^{-1} \cdot K^{-1}$
ρ_f	density of PU foam	kg·m⁻³
ρ_s	density of PU polymer	kg·m ⁻³

The PET foam cell structure is somewhat different from that of the PUR foam, probably due to the fact that the foaming processes are different. The PUR foam is formed in an exothermal reaction between a polyol and an isocyanate while the PET foam is a thermoplastic material produced in an extrusion process.

The cell size and the distribution of polymer between struts and windows differs between the foam types (geometry factor). Since the PET foaming process is under development it is not possible to say if the same value of f_s , that has been used for the PUR foam also is valid for the PET foam.

Radiation

The thermal conductivity due to radiation in the foam takes place in all directions from the surface of the cells, with the net energy transfer from hot to cold side. The radiation is strongly dependent on temperature. Other factors with influence are cell structure, size of cells and radiative properties of the cell walls.

For the PET foams studied the average cell size is larger than that of the PUR foams used for district heating pipes. The average cell size for the PET foam was about 0.6-1.0 mm depending on density.

The radiation between the cell walls of PUR foam can be calculated by the Rossland equation with the extinction coefficient of the cell wall material $K_w = 60000 \text{ m}^{-1}$ suggested by Glicksman [1994].

$$\lambda_{radiation} = \frac{16 \cdot \sigma \cdot T^3}{3 \cdot K} \qquad \text{where} \qquad K = 4.10 \cdot \frac{\sqrt{\frac{f_s \cdot \rho_f}{\rho_s}}}{d} + \left[\frac{(1 - f_s) \cdot \rho_f}{\rho_s}\right] \cdot K_w$$

Symbols:

K	extinction coefficient	m^{-1}
Т	temperature	Κ
d	cell diameter	m
f_s	fraction of solid in struts	-
$ ho_f$	density of PU foam	kg·m ⁻
$ ho_s$	density of PU polymer	kg·m ⁻³
σ	Stephan Boltzman constant = $5.7 \cdot 10^{-6}$	W·m ⁻² ·K ⁻⁴

The Rossland equation can most likely also be used for the PET foam. However the value of the extinction coefficient is not known. Due to the fact that very little research work has been reported on the PET foam properties it is not possible at this stage to make any statement on the exact radiation properties of PET foam.

2.3.2 Measurements

The experimental work to establish the thermal conductivity and the material characteristics influencing the thermal conductivity was done at Chalmers University of Technology. The thermal conductivity was determined in a Heat Flow Apparatus according to ISO8301.



The cell gas content was determined by a gas chromatographic method developed by Ramnäs [1995].





Figure 31. Equipment for cell gas analysis. A cylindrical foam sample, PET or PUR, diameter 20 mm, is grind with a rotating steel knife in a metal cylinder (upper part of the Figure) and the released cell gases are collected in a gas tight glass syringe (middle part). The volume of the released gases is determined and the gas mixture is analysed in a gas chromatograph (lower part). When the research project started only PET foam boards blown with HCFCs and HFCs were available. The insulation properties of this "first generation" of PET foam were determined in order to adapt the sampling and measurement techniques to the PET foam. The results are shown in Table 5. Even if environmentally unacceptable blowing agents were used and the densities were too high for our purposes, the thermal conductivity measurements gave a clear indication that PET foam with a cell diameter and density corresponding to a PUR foam would have a thermal conductivity of the same magnitude as a PUR foam.

Next phase was to eliminate HCFCs and HFCs. The blowing agent for this "second generation" foam was carbon dioxide. However, it was difficult to reduce the density below 100 kg·m⁻³. A very successful development phase resulted in a "third generation" cyclopentane blown PET foam with densities corresponding to those used for PUR insulated district heating pipes.

PET foam	Blowing agent	Air	Foam	Thermal
blown with	partial pressure	partial pressure	density	conductivity
	kPa	kPa	kg∙m⁻³	$W \cdot m^{-1} \cdot K^{-1}$
	First generat	ion of PET-foam		
HFC-152a	76	3	120	0.028 @20°C
HCFC-142b/HCFC-22	66^*	37	91	0.030 @20°C
				-
HFC-152a	54	21	156	0.035 @20°C
	Second gen	eration of PET foam		
Carbon dioxide	0.2	19	144	0.043 @20°C
				Ŭ
	Third genera	tion of PET foam		
Cvclopentane	58	6.4	55	0.028 @ 40°C
J 1	-		-	0

* Volume ratio HCFC-142b/HCFC-22: 61/39

The total cell gas pressure and the cell gas content of the cyclopentane blown PET foam have been determined (Table 6).

<i>Table 6. Cell gas content (vol-%) of</i> <i>a cyclopentane blown PET foam.</i> <i>Foam density 55 kg·m⁻³ Total cell</i>	Oxygen	Nitrogen	Ca dic
gas pressure 68 kPa (@24°C).	2.0	7.4	(

Oxygen	Nitrogen	Carbon dioxide	Cyclopentane	Water	Others
2.0	7.4	0.4	85	3.9	1.5

Table 5. Characteristics of the analysed PET foams

gas

2.3.3 Calculations

When calculating the thermal conductivity a number of material parameters have to be known. Some of the parameters needed for the calculation are difficult to determine and the calculation model can only describe the foam geometry and structure in rough outlines. However, the calculated thermal conductivities of two PET foams are presented in Table 7. These foams are earlier mentioned as foam of the "second" and "third generation" (Table 5).

For the originally carbon dioxide blown PET foam it should be noticed that the blowing agent, carbon dioxide, has diffused out of the foam and that only air is present in the cells. The calculation of the total thermal conductivity gives $0.043 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ and the same value was obtained from the measurements. The fraction of solids in the struts (f_s) is assumed to be 0.4 (f_s =1 indicates that all polymer material is located in the struts and f_s =0 that all polymer material is located in the windows).

The cell gas in the originally cyclopentane blown PET foam contained 11 vol-% air and 89 vol-% cyclopentane. The calculated thermal conductivity (0.0276) agrees almost exactly with the measured value (0.028).

	Cell gas: air Foam density: 144 kg·m ⁻³ Thermal conductivity @ 20 °C W·m ⁻¹ ·K ⁻¹	Cell gas: cyclopentane/air Foam density: 55 kg·m ⁻³ Thermal conductivity @ 40 °C W·m ⁻¹ ·K ⁻¹
$\lambda_{gasmixture}$ $\lambda_{polymer}$ $\lambda_{radiation}$	0.0258 0.0147 0.0026	0.0161 0.0054 0.0061
λ_{total}	0.0431	0.0276

The thermal conductivity of a typical newly produced cyclopentane/carbon dioxide blown PUR foam for district heating pipes (density about 60 kg·m⁻³) is about 0.027-0.028 W·m⁻¹·K⁻¹ at 50°C. The contribution of radiation and conduction in the polymer to the thermal conductivity of insulating foams is constant over time. It is estimated at 0.010 W·m⁻¹·K⁻¹ @ 50°C for a typical PUR foam used for district heating pipes [Olsson 2001].

Table 7. Calculated values of the different contributions to and the total thermal conductivity of two PET foams.

Since the thermal conductivity strongly depends on the temperature it is important to establish this relation. For five newly manufactured PUR foam boards with excellent insulating performance the thermal conductivities have been determined at various temperatures. Due to variation in density, cell size and structure and cell gas content there is a difference in thermal conductivity between the foams. However, the dependency of temperature on the thermal conductivity seems to be the same for all foams within the temperature range 10-40°C, see Figure 32 [Jarfelt 2006]. The temperature dependence for PET foam is likely to be similar to PUR foam.





2.3.4 Long term thermal performance

In order to describe the thermal performance of an insulating foam, both the thermal conductivity of the new foam and the aging behaviour of the foam must be known. The aging behaviour is often called long term thermal performance (LTTP) and depends upon the fact the cell gases will diffuse out of and air into the foam until equilibrium is reached. The cell gas composition will change over time. Since most blowing agents have lower thermal conductivities and diffuse slower in the foam than air, this means that total thermal conductivity of the foam will increase over time. Thus, a foam having low thermal conductivity when it is tested directly after production, may not necessarily be the best choice because of its unfavourable aging behaviour.

The cell gases are partly dissolved in the polymer matrix. For many foams about 50% of the total amount of gas in the foam is dissolved in the polymer. When the cell gas diffuses out of the foam at a certain rate, the concentration of gas in the cells may decrease much slower since the gas dissolved in the polymer will desorb to the gas phase in the cells. The dissolved gas will act as a buffer decreasing the rate of loss of insulation performance of the foam. Thus, the change of insulation performance of a foam depends both on the diffusion characteristics of a gas in the polymer matrix and on the solubility of the gas in the polymer matrix. The diffusion process can be described by the diffusion coefficient. If both the diffusion and the solubility are considered, the change of concentration of a cell gas can be described by the effective diffusion coefficient (D_{eff}).

If the effective diffusion coefficients of the different cell gases in the foam are known the long term thermal performance of the foam can be predicted.

Determination of the effective diffusion coefficient

In order to determine the effective diffusion coefficients cylinders of PET foam were stored at different temperatures. In order to prevent unwanted longitudinal mass transport of cell gases, plates of aluminium were glued with epoxy onto the ends of the cylinders. The foam samples were analysed after different storage times at the respective temperatures. The partial pressures of carbon dioxide, oxygen, nitrogen and cyclopentane were determined after crushing the foam, measuring the total volume of the released cell gases and analysing the cell gas composition by gas chromatography [Ramnäs 1995]. The change of cell gas pressure in the cylinders was assumed to follow Fick's law. The mean partial pressure for each gas can be calculated according to an equation for cylinders with only radial diffusion.

$\overline{p}_i(t) = p_i(t)$	$p_{0i} \sum_{j=1}^{\infty} \frac{4 \cdot \exp(-\frac{1}{2})}{2}$	$\frac{\left(-\beta_{0j}^{2}\cdot D_{e\!f\!f,i}\cdot t\right)}{\left(\beta_{0j}\cdot a\right)^{2}}$	
where	\overline{p}_i	mean partial pressure of component i	(kPa)
	p_{0i}	Initial partial pressure of component i	(kPa)
	$D_{eff,i}$	effective diffusion coefficient of component <i>i</i>	$(m^2 \cdot s^{-1})$
	$\beta_{0,i} \cdot a$	root of the zero order Bessel function	
	a	cylinder radius	(m)
	t	time	(s)

The results from tests on four different PET foams are shown in Table 8. The HCFC and HFC foams were already in production and used for other applications than insulation purposes when the project started. However, some process changes were made in order to produce PET foam boards with a geometry suitable for our testing purposes. Foams of the second and third generation of carbon dioxide and cyclopentane blown PET foams have also been included in the study of the effective diffusion coefficients. The result showed that the diffusion coefficients for the air components were about one tenth of those for diffusion in a PUR foam. Consequently the PET foam will exhibit a better long term thermal performance than the PUR foam.

Density (kg·m ³)	91	120	120	157	157	157	157	55
Blowing agent	HCFC- 142b/22	HFC- 152a	HFC- 152a	CO ₂	CO ₂	CO ₂	CO ₂	Cyclo- pentane
Temperature (C°)	23	23	60	23	40	60	90	80
$\begin{array}{c} D_{eff}(nitrogen) \\ 10^{-13} \cdot m^2 \cdot s^{-1} \end{array}$	2.7-3.0	3.0	16	1.5	3.5	7.0	70	60
$\frac{D_{eff}(oxygen)}{10^{-13}} \cdot m^2 \cdot s^{-1}$	-	20	95	15	20	40	290	220
$\begin{array}{c} D_{eff}(carbon \\ dioxide) \\ 10^{-13} \cdot m^2 \cdot s^{-1} \end{array}$	-	-	-	45	85	150	1000	-

Table 8. The effective diffusion coefficients $(D_{eff} \cdot 10^{-13} \text{ m}^2 \cdot \text{s}^{-1})$ of gases in PET foam with different densities and blowing agents.

The temperature dependence of the effective diffusion coefficient can be described by the activation energy. Using the values of the effective diffusion coefficients at different temperatures and assuming an Arrhenius type relationship the activation energy can be calculated.

$$D_{eff} = D_0 \cdot \exp\left[\frac{-E_D}{R \cdot T}\right]$$

where

D_0	pre-exponential factor	(-)
E_D	activation energy	$(J \cdot mole^{-1})$
R	the gas constant $= 8.314$	$(J \cdot K^{-1} \cdot mole^{-1})$
Т	temperature	(K)

The activation energy for oxygen, nitrogen and carbon dioxide has been calculated based on the result from the PET-foam blown with carbon dioxide.

Nitrogen	Oxygen	Carbon dioxide	Cyclopentane
3.2·10 ⁴	2.5·10 ⁴	$2.5 \cdot 10^4$	-

The density has an influence on the diffusion properties. High density often results in reduced diffusion rate due to thicker cell walls (windows). To the extensive experimental study conducted on the carbon dioxide blown foam with a density around

140 kg·m⁻³, a minor study on the "third generation" of cyclopentane blown PET foam with a density of 55 kg·m⁻³ was added. The effective diffusion coefficients for nitrogen and oxygen in foams of very different densities are shown in Figures 33 and 34.

Table 9. Activation energy E_D (J·mole⁻¹) for gases in PET foam (carbon dioxide blown).









Table 10. Effective diffusioncoefficients of nitrogen and oxygenin carbon dioxide blown(foam density: 144 kg·m³) andcyclopentane blown (foam density:55 kg·m³) PET foams.The results are calculated assumingan Arrhenius relationship.

	Effective diffusion coefficient D _{eff} ·10 ⁻¹³ (m ² ·s ⁻¹) of nitrogen and oxygen in PET foam					
	20°C	40°C	60°C	80°C		
	Foam density 144 kg·m ⁻³					
nitrogen	1.5	3.8	8.4	17		
oxygen	13	25	45	74		
	Foam density 55 kg·m ⁻³					
nitrogen	5.2	13	29	60		
oxygen	38	74	130	220		

In Table 10 it is shown that more solid material in the foam structure will slow down the diffusion. The magnitude of influence will probably depend on a number of reasons, i.e. distribution of polymer material between struts and windows, cell size and cell geometry and uniformity of foam structure. In this report two PET foams with quite different densities, 144 kg· m⁻³ and 55 kg·m⁻³, have been studied. The ratio of the effective diffusion coefficient is 1:3.4 for nitrogen and 1:2.9 for oxygen which roughly corresponds to the ratio of the densities.

2.4 Environmental performance

2.4.1 Introduction

Proposed new technical solutions should be studied regarding environmental performance before taken into use. New technology should generally not be employed if it has worse environmental impact compared to the present solution. Environmental life cycle assessment is one method to describe the performance of a technical solution, which has been used in this study to benchmark the PET insulated district heating pipes compared to PUR insulated district heating pipes.

Environmental impacts in a life cycle perspective

The concern for environmental issues was for a long period of time concentrated to different forms of point sources, e.g. chimneys at factories or exhaust gases from vehicles. Step by step, the understanding of the impacts from society on ecosystems has broadened. It is not only the production or the use of a specific item that causes impacts on the environment, but rather the whole chain of processes needed to give the function of a specific product, such as extraction and transports of raw materials, upgrading of raw materials, the actual production of the product, necessary energy transformations and waste collection and treatment (Figures 30 and 31).

This form of life cycle thinking was put into focus through the publishing of Our Common Future [Brundtland Commission, 1987], the Brundtland commission report to the UN environmental meeting in Rio de Janeiro 1992. To be able to reach the goal of sustainable development, it is necessary to produce products and services with high ecoefficiency and lowest possible total life cycle emissions and environmental load.

Life cycle assessment (LCA) is a way to calculate and evaluate, quantitatively, the environmental load that is caused by a product or service during all phases of its life cycle, see Figure 32. The environmental impacts from each of the phases of the product's life are investigated and summed up into a total for the life cycle. Environmental impact is generally interpreted as emissions of different pollutants and wastes as well as consumption of natural resources and consequences of the extraction. It is, in principal, possible – but difficult and seldom practiced – to include other parameters, such as noise and working environment.



Figure 35. Traditionally, environmental work has been focused on emissions from point sources, e.g. from the chimney of a factory (modification of figure from Hauschild et al, 1998) Figure 36. A product or process studied in an environmental life cycle perspective; all stages in the production process are taken into account (modification of figure from Hauschild et al, 1998)



Figure 37. A simplified illustration of a life cycle system for a product or a service. Natural resource consumption, emissions, etc. are summarized for all the parts of the system into a total environmental load



To be able to perform the inventory of environmental impacts, it is necessary to decide what shall be included in the system under study. This is done in the first phase of an LCA, denoted Definition of Goal and Scope. Within this phase the system boundaries are set up and a functional unit to be studied is chosen. The second phase is the Inventory Analysis, the collection and compilation of the environmental impacts caused by the different activities included in the system. For each environmental impact, a total is calculated for the system. The result is a list with all the different parameters of natural resource consumption and emissions studied. This list is denoted inventory result (or eco-profile). If the study is ended here, it is called a Life cycle inventory (LCI). If the study is taken further and the result is interpreted, the study is called a life cycle assessment (LCA).

Hence, LCA is a way to give a holistic view of the combined environmental impact from all phases of the life cycle of a product or a service; the environmental impact from "cradle to grave". It is important to remember that the result of an LCA is totally dependent on what is included in the system under study as well as how the study has been performed. To fully understand the result from an LCA study it is necessary to read in detail and understand the goal and the scope chosen for the specific LCA study, and thus very important that LCA reports contain extensive information on goal and scope as well as data for the included activities - if not, the results from the study will not be possible to understand. The existing standards and "codes of praxis" for LCA have all mainly been set up to guarantee that studies are performed and reported in such a way that interpretation of the result is possible for an external reader.

International standard for environmental management and LCA.

The Standard ISO 14040 describes a methodology for performing an LCA in a systematic way [ISO 14040]. This standard constitutes one part of the series of standards on environmental management, the ISO 14000 series, also including standards for environmental management systems (ISO 14001), environmental auditing (ISO 14010), environmental labeling (ISO 14020) and environmental product declaration (ISO 14025).

The ISO standard for life cycle assessment describes a structure to follow when conducting the study, Figure 33. If the result from an LCA is to be used externally for product information or advertisement, the standard demands a critical review of the study by a body independent of both the practitioner and the commissioner.



In the definition of the goal and scope, the ultimate purpose of the study shall be chosen and stated. Here, the functional unit, the base/basis/common denominator for the calculations, shall be established, and methodological choices that influence the subsequent work shall be declared. System boundaries shall be defined, e.g. geographic boundaries and boundaries in time. An example (of course simplified) could be that "the product is manufactured in northeastern USA, using modern production technology; the product is subsequently exported to Sweden where it is sold, used and ultimately treated in the Swedish waste management system."

There are different forms of LCA studies. A study can describe a specific case, where the production of a product in a specific factory at a specific time is investigated, and using data for this specific production case. A study can also be more general, using general or averaged data for e.g. the average production of polyethylene plastics from 20 large manufacturers in Europe where different producers within the branch use different production technologies. An LCA study can also describe a scenario where best available data is used to describe a production that does not yet exist, to create a basis for managerial or political decisions.

An LCA study may in principle be qualitative and describe the environmental impact from the system under study in words. However, the more common case is that LCA studies are quantitative

Figure 38. The structure for performing a life cycle assessment according to ISO 14040. The double arrows indicate the iterative structure of a life cycle study and present, as far as possible, figures for the environmental impacts from the different parts of the LCA system.

After the goal and the scope of an LCA have been defined and the system boundaries decided, the inventory part of the study starts. In the inventory analysis, data is collected from the different activities within the system boundaries, and for each environmental impact, the collected information is summed up for the whole LCA system. Some information can be found in databases and in literature. Process data are sometimes available for a few activities, but the farther away from the actual process or product under study that you get in the life cycle, the more difficult is normally the gathering of data of good quality. This is generally the most time consuming part of the study.

In order to facilitate the life cycle impact assessment, the inventory analysis data can be grouped in different ways. One method is to group together all inventory parameters (emitted substances etc.) that contribute to a specific environmental theme, e.g. all emissions found during the inventory that contribute to acidification, ozone depletion or global warming, the use of finite resources and so on. To group the data from the inventory analysis in this way is denoted classification.

If all impacts in one classification group are expressed in a common unit, it is possible to add the impacts together and obtain one single value for the total contribution from the different impacts. All emitted substances within the category "global warming" can e.g. be expressed in carbon dioxide equivalents and summed into the life cycle contribution to global warming. In the same way, the amounts of emitted substances contributing to acidification can be expressed in hydrogen ion equivalents (or sulfur dioxide equivalents) and summed up into the life cycle contribution to acidification; ozone depleting substances can be expressed in CFC-11 equivalents and so on. These kinds of recalculations are called characterizations of the life cycle inventory result.

In practice, LCA is partially an iterative method. Generally, it is not until a first impact assessment has been performed that it is possible to tell which parts of the system that contribute most severely to the environmental impacts of the system, and hence need special attention during the inventory analysis. Consequently, it might be necessary to go back and redo parts of the inventory work. During the work with the LCA study it might even become clear that the initial goal definition was not clear or complete, making it necessary to go back and make adjustments or clarifications. This is the reason for the double arrows in Figure 33.

2.4.2 Environmental comparison of PET and PUR foam insulated district heating pipes

The aim of this study is to investigate the environmental performance of DH pipes insulated with PET foam (polyethylene terephthalate, carbon dioxide blown, virgin or recycled PET) and compare with pipes insulated with PUR foam (polyurethane, cyclopentane/carbon dioxide blown). The focus is on pipe production and network use; installation of the district heat distribution system is assumed to be similar in the two cases. The work is based on previous LCA-studies on PUR foam insulated DH pipes [Persson et al, 2005; Fröling, et al. 2004; Fröling and Svanström, 2004] and studies of the long-term thermal performance of PET foam described earlier in this report and also in [Mangs et al, 2004] and [Mangs et a, 2005]. Since commercially viable methods to produce PET foam insulated DH pipes are not yet available this is a theoretical study of environmental feasibility.

The environmental comparison was performed using life cycle assessment (LCA) methodology. The life cycle of a DH pipe can be divided into "pipe production", "network construction", "network use" and "post-use handling" see Figure 4. Few district heating networks have to date been taken out of use, thus post-use management has not been included in this study. From an environmental perspective, the entire life-cycle of the pipes: pipe production, network construction, network use and post-use handling (Figure 34) [Fröling 2002, Mangs 2005].





System description and inventory

Single DH pipes of the DN100/225 dimension with a casing thickness of 3.4 mm were studied [EN253:2003.]. The steel pipes are 12 m long, of which about 20 cm at each end is left uninsulated to facilitate installation welding. The functional unit – to which the results refer – is 1 meter of the distribution system (including 1 m flow pipe and 1 m return pipe) and its use over a period of 30 years. The inventory results were characterized as global warming potential (GWP) given in kg CO_2 -equivalents, acidification potential (AP) in kg SO_2 -equivalents and depletion of finite resources (RD) in kg·year⁻¹ (current annual global consumption of a finite resource divided by the estimated reserves of the same resource) [Nord, 1995; Hauschild and Wentzel, 1998].

The six different types of DH pipes studied are shown in Table 11. Pipes insulated with two PET foam densities and with virgin and recycled raw material were studied and compared with PUR foam insulated pipes with two alternative blowing agents. All data on PET foam insulated DH pipes are preliminary, as such pipes are not commercially produced.

At the start of the environmental part of this study it was not possible to produce cyclopentane PET foam. Thus, only carbon dioxide blown foam has been considered in this part of the report. High density PET foam was chosen, due to the fact that it is the only foam that has so far undergone detailed investigation in terms of long-term thermal performance [Mangs et al. 2005]. The pipe insulated with PUR foam blown with cyclopentane/carbon dioxide represents an insulating foam used today. Purely carbon dioxide blown PUR foam is not frequently used in DH pipes of the studied dimension, but is included here in order to benchmark towards the carbon dioxide blown PET foams. Both studied types of PUR foam insulated pipes have casings made of high density polyethylene (HDPE), while the PET foam insulated pipes are assumed to have PET casings, which will facilitate material recycling of the pipes when taken out of use.

Table 11. The different DH pipes included in the study	Pipe alternatives
	DLID (arr)

Pipe alternatives	Foam type	Foam density (kg·m ⁻³)	Blowing agent	Casing material
PUR (cp)	PUR	86	Cyclopentane/ CO ₂	HDPE
PUR (CO ₂)	PUR	77	CO ₂	HDPE
PET (HD,virgin)	virgin PET	157	CO_2	virgin PET
PET (HD, recycl)	recycl PET	157	CO_2	recycled PET
PET (LD, virgin)	virgin PET	86	CO ₂	virgin PET
PET (LD, recycl)	recycl PET	86	CO_2	recycled PET



District heating pipe production

A summary of the activities included in the production of the pipes is presented in Figure 35. Inventory data for the following parts of the production process were assumed to be identical for PUR and PET foam insulated pipes: steel pipe, copper wire and energy used in the factory (e.g. for pipe assembly, lighting and heating of the factory buildings).

The production of all pipe variants is assumed to take place in Göteborg, Sweden. The inventory data on cyclopentane/carbon dioxide blown PUR foam insulated DN100 pipes are described in detail in [Fröling et al., 2004]. For pipes insulated with carbon dioxide blown PUR foam, modifications were made in accordance with information provided by [Carlsson, 2002].

Recycling of PET was approximated with data from a study on recycling of polyethylene packaging in Sweden [Powerpipe Systems AB, 2005]. For the case of PET foam made from recycled polymer, all the emissions and energy used in the recycling process were ascribed to the PET foam production, while all impacts from extraction of virgin PET and bottle production were ascribed to the PET bottles.

Figure 35. Summary of the LCA activities considered for production of PET and PUR foam insulated DH pipes.

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Some minor materials were omitted in the study of PET foam insulated pipes, in view of the fact that a previous study of PUR foam insulated pipes showed that the production of the main materials (steel pipe, foam insulation, casing pipe and copper wire) gives rise to the major part (90 % or more) of the environmental impact [Fröling et al., 2004].

Inventory data for virgin PET granule production were taken from an LCA performed by APME [Bousted,2002]. Information about the production of carbon dioxide blown PET foam was provided by the pilot plant run by B.C. Foam in Italy [B.C. Foam, 2005]. The figures used pertain to the production of foam boards, but it is assumed that the amount of energy and material would be similar for the production of foam for pipe insulation. The energy required to extrude 1 kg carbon dioxide blown PET foam board is 1 kWh. Talcum, a nucleating agent that enhances the foaming process is added (0.5 % by mass) to the PET granules before they enter the extruder. Data for talcum production were provided by a Swedish talcum producer [Handöls Täljstens AB, 2005]. The blowing agent, carbon dioxide is assumed to be a by-product thus only the energy needed to compress the gas, 0.28 kWh electricity per kg of carbon dioxide, was taken into account [AGA Gas, 2005].

Network construction

Laying of PET and PUR foam insulated DH pipes is assumed to have the same environmental impact. Inventory data for construction of a district heat distribution network built of PUR foam insulated DH pipes in urban and green areas were applied for both PET and PUR foam insulated pipes [Fröling and Svanström, 2004]. It was assumed there is not significant environmental difference in laying the tow types of pipes.

Network use

During network use environmental impacts from heat generation to compensate for heat losses have been considered. Heat losses from DH networks increase over time, as the thermal conductivity of the insulating foam increases due to diffusion of air into and blowing agent out of the foam. The average thermal conductivities of the foams and the total heat losses from the DH networks over a period of 30 years were calculated according to a method described in [Persson and Claesson, 2005]. The transport coefficients used to calculate the change in the cell gas mixture of the foam are given in Table 12 [Mangs et al, 2005; Svanström et al., 1999; Swedish District heating Association, 2005]. The flow and the return pipe annual temperatures averages were estimated to 80°C and 40°C, respectively. The average temperature of the soil surrounding the pipes was assumed to be constant at 15°C (average over the year).

		Initial partial pressure (kPa)			
		$CP CO_2 O_2 N_2$			N_2
PUR foam CP/CO ₂ -blown	25°C	42.4*	66.5	0.4	0.7
PUR foam CO ₂ -blown	20°C	-	122	0.2	0.7
PUR foam CO ₂ -blown [#]	20°C	-	80	2.5	2.5

The same initial partial pressures are assumed for all PET foam types.

* 20 % by mass of the total cyclopentane content (gas + liquid) as liquid in the foam.

Table 12. Initial partial pressures of cell gases at 20-25°C for each of the studied foam types.

CP = cyclopentane $CO_2 = carbon \ dioxide$ $O_2 = oxygen$ $N_2 = nitrogen$ The thermal conductivity of the foam is the sum of conduction through the cell gas (λ gas), conduction through the solid polymer (λ pol) and radiation (λ rad). Wassiljeva's equation as modified by Mason and Saxena was used to calculate λ gas [Reid et al, 1977]. For the PUR foams and the low density PET foam, λ pol was calculated with the aid of a matrix conduction model [Nielsen, 1998] and λ rad according to the Rossland equation [Siegel and Howell, 2001]. An equation from [Mangs et al, 2005] was used to calculate the sum of λ pol and λ rad for the high density PET foam. The resulting average thermal conductivities of the studied pipe alternatives (λ pipe) over 30 years of use are reported in Table 13.

Pipe alternative	λ_{pipe} (mV	$W \cdot m^{-1} \cdot K^{-1}$
1	Flow pipe	Return pipe
PUR (CP)	29.9	27.4
PUR (CO ₂)	37.6	34.9
PET (HD, vir)	39.1	36.1
PET (HD, rec)	39.1	36.1
PET (LD, vir)	30.1	28.0
PET (LD, rec)	30.1	28.0

Table 13. Average thermal conductivities (λpipe) for each pipe alternative over 30 years of use.

Average Swedish district heating fuel mix in 2000 (32 % renewable fuel, 29 % waste incineration, 15 % heat pumps, 6 % oil, 5 % peat, 5 % natural gas, 4 % coal and 4 % electricity) was used to convert the calculated heat losses into environmental impacts [Swedish District Heating Association, 2000]. Natural gas combustion (heat only boilers) was one of the major primary energy source for district heat in several European counties in 2001 and was therefore included as a comparison [Euroheat & Power, 2003].

2.4.3 Results and discussion

The characterized environmental impacts for the six studied DH pipe alternatives are shown in Table 14. The largest environmental impacts during the studied parts of the life-cycle of these pipes are attributed to heat losses during network use. The contribution of this phase to the whole life cycle is 59-97 % for all impact categories. During the use phase, heat generation with natural gas boilers gives rise to about half of the acidification potential, 3 times higher global warming potential and 5 times higher resource depletion compared to the average Swedish heat mix.

Table 14. Characterised environmental impacts from the first three phases of the life cycle of a 1 m DN100 pipe network (1m flow pipe and 1 m return pipe) over 30 years of use in terms of global warming potential (GWP, kg CO_2 -equivalents), acidification potential (AP, kg SO₂-equivalents) and resource depletion (RD, kg-year⁻¹)

	Pipe pr	Network construction*Network use, 30 yearsPipe production(green area / urban area)(Swedish fuel mix/national mix/		Network construction* (green area / urban area)		s tural gas)			
Pipe alternative	GWP	AP	RD	GWP	AP	RD	GWP	AP	RD
PUR (CP)	63	0.31	0.65	22/39	0.20/0.37	0.20/0.34	500/1500	2.7/1.1	1.7/8.4
PUR (CO ₂)	62	0.30	0.64	22/39	0.20/0.37	0.20/0.34	620/1900	3.3/1.3	2.2/10.6
PET (HD, vir)	81	0.52	0.69	22/39	0.20/0.37	0.20/0.34	650/2000	3.5/1.4	2.3/11.0
PET (HD, rec)	41	0.11	0.32	22/39	0.20/0.37	0.20/0.34	650/2000	3.5/1.4	2.3/11.0
PET (LD, vir)	67	0.38	0.56	22/39	0.20/0.37	0.20/0.34	500/1500	2.7/1.1	1.8/8.5
PET (LD, rec)	41	0.10	0.32	22/39	0.20/0.37	0.20/0.34	500/1500	2.7/1.1	1.8/8.5

*The network construction results are mainly influenced by excavation work and transports, and are thus similar for all studied pipes, as all pipes have the same dimension and approximately the same weight per meter.

District heating pipe production

The environmental impacts for pipe production are illustrated in Figure 36. The choice of blowing agent in the PUR foam does not result in any significant difference in any of the impact categories during the production phase. The difference between the pipe alternatives is mainly associated with the choice of insulation material. All other materials and activities included in pipe production give rise to about the same impacts, regardless of the type of pipe. The pipe with virgin high density PET foam insulation has the highest environmental impacts compared to the other alternatives, which is attributed to the large amount of insulation material. If the density were lowered, the amount of PET would be reduced and the impacts similar to that of the PUR foam insulated pipes. Significant reduction in both emissions and resource use could be achieved by using recycled PET.



Figure 36. Characterised environmental impacts of pipe production: normalised values [PET(HD,vir) = 1] of global warming potential, acidification potential, and resource depletion for the six studied types of pipes. Recycled material shows significantly lower environmental impact

Pipe production

Network construction

The network installation procedure is modelled in the same way for all the six pipe alternatives. Although some new methods may have to be developed to install PET foam insulated pipes, the excavation, transport of materials etc. are not expected to differ much from the construction of traditional pipe networks. In a study of the environmental impacts of network construction, these activities contributed 85 % or more [Fröling and Svanström, 2004]. In urban areas deeper trenches and restoration of asphalt are necessary, which leads to higher values in all impact categories compared to green areas (Table 14).

Network use

The heat losses during use are compared in Figure 37. The results are based on the simulated average heat losses over 30 years. The simulated heat losses are not dependent on whether virgin or recycled material is used. The results also illustrate the fact that gases generally diffuse more slowly in PET foam compared to PUR foam.

Pipes insulated with carbon dioxide blown high density PET foam and carbon dioxide blown PUR foam give rise to similar impacts. It is possible to achieve a similar environmental performance to that of cyclopentane blown PUR foam using lower density PET foam. DH pipes insulated with low density PET foam blown with other environmentally friendly blowing agents with low thermal conductivity and slow diffusion properties could even be superior to pipes insulated with PUR foam. This project has shown that it is possible to produce low density cyclopentane blown PET-foam, but methods generating cell seizes appropriate to district heating pipe insulation is still under development.



Network use

Figure 38 a-c shows the environmental impacts characterised as GWP (a), AP (b) and RD (c) during the use phase for the Swedish heat mix and natural gas boilers as heat source. Due to the high amount of renewable fuels used for Swedish district heat production (32 %), the carbon dioxide emissions are about one third of those of the natural gas boiler system. The acidifying effect is, however higher in the Swedish case, since the heat mix contains small proportions of coal and peat (4 and 5 % respectively). A system based exclusively on fossil fuels result in a resource depletion that is almost five times higher than the Swedish heat mix.

Figure 37. Heat losses of a 1 m DN100/225 pipe network (1m flow pipe and 1 m return pipe) over 30 years: Normalized values, PET(HD) = 1. All PET alternatives use carbon dioxide as insulating gas. Figure 38. Characterized environmental impacts from the use phase of a 1 m DN100/225 pipe network (1m flow pipe and 1 m return pipe) over a 30-year period with regard to

- a) global warming potential (GWP),
- **b**) acidification potential (AP) and
- *c*) resource depletion (RD)



PUR (365) PUR (CO2)

PET (HD)

PET (LD)

0.0

PUR (CP)

Combined results from the three life cycle phases

Figure 39 a-c shows the environmental impacts from the three life cycle phases: pipe production, network construction in urban areas and impacts due to heat losses based on the average Swedish district heat mix. DH pipes insulated with low density, carbon dioxide blown PET foam from virgin resin have the potential to perform as well in environmental terms as cyclopentane blown PUR foam insulated pipes. If recycled PET even lower environmental impacts than those of traditional pipes may be achieved. These results can be validated when commercial production methods for pipes insulated with low density PET foams blown with environmentally friendly insulating gases have been developed. The potential for improvement in the long-term thermal and environmental performance of PET foam is probably high, in view of the fact that it is a new material.

The relative contribution from the heat losses to the whole life cycle depends on the type of heat source that is used to produce the heat. Since natural gas is a non-renewable resource, it has a greater impact on global warming and resource use, compared to the average Swedish district heating mix, which utilizes 32 % renewable fuels. The higher acidification potential of the Swedish district heating mix is to a large extent due to the presence of coal (4 %) and peat (5 %) in the fuel mix, which result in acidifying emissions of sulphur dioxide.

3. Discussion / Comparison of PET and PUR foam

3.1 Mechanical performance

The mechanical performance has been studied using the European standard EN 253:2003 as the reference. This standard is written with respect to PUR foams for pre-insulated district heating pipes. The standard applies for continuous operation at 120 °C for 30 years. However, for pipes of small dimensions, outer diameter \leq 120 mm, both temperatures and mechanical loads are lower.

Compressive strength

The short-term compressive strength decreases with increasing temperature with a drastic change at approximately 80°C. This is due to the material's glass transition temperature. It can be seen that the compressive strength at high temperatures does not fulfil the requirements of 0.3 MPa for the PUR foam according to EN 253. It should be kept in mind, however, that this requirement is likely unnecessarily conservative for small pipes.

The short-term elastic modulus behaves in a similar fashion, with obvious temperature dependence and a significant drop of stiffness around the glass transition temperature.

Water permeability and vapour resistance

The tested PET foams were impermeable to liquid water and only vapour diffused through the PET foam. The vapour diffusion coefficient was $5 \cdot 10^{-8} \text{ m}^2 \cdot \text{s}^{-1}$. That means that the vapour resistance was approximately 10 times greater for the PET foam than for a regular PUR foam.

Water absorption

The water absorption for PET foam was found to be 7 %- vol. The requirement on PUR foam in EN 253 is a maximum of water absorption of 10 %. PUR foam usually absorbs around 5 %.

Glass transition temperature

The results verify that the PET foams turn softer at approximately 80 °C, as could also be seen from the short-term compressive strength measurements at different temperatures. It is not possible to make a direct comparison with PUR. PUR is a thermoset material which does not undergo this kind of phase transition. However, measurements on carbon dioxide blown PUR foam by thermomechanical analysis (TMA) showed a softening temperature around 110-120°C [Jarfelt, 1992].

Creep behaviour

The creep properties of PET foam seem to be very good. Extrapolation of the creep curves to 30 years of technical service does not indicate any significant creep deformation. Furthermore, the creep rate at high temperature is well in line with PUR foam.

Flexibility – bending properties

Samples of PET taken parallel to the extrusion direction are much stiffer and much more brittle, while samples taken parallel to the cross direction are so flexible that no fracture was seen up to the testing limit of 12 % strain. The material can easily be processed to withstand bending strains to a sufficient degree for flexible district heating pipes.

The PUR foams behave in a little more brittle manner. The "flexible" foam did not exhibit a significantly more flexible behaviour than the rigid foam. A strain limit of approximately 11 % was seen for both materials. It is also interesting to note that the flexible foam is much stiffer than the rigid foam.

3.2 Insulating performance

Polymer matrix

The thermal conductivity of solid PUR polymer from district heating pipes was reported to be $0.21 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ @ 23°C with a linear increase over temperature of 0.2 mW·m⁻¹·K⁻¹ [Nielsen 2000]. Literature values of the thermal conductivity of solid PET polymer are similar to PUR. The type of PET (molecular weight and structure) and the degree of crystallinity will only affect the solid thermal conductivity to a minor degree.

Radiation

The PET foams studied show a cell structure with larger cells than those normally obtained for PUR foams. The cell size of the PET-foams varies between 0.6 to 1.0 mm. At present the production process is not optimized in the respect of minimizing the cell size. Two types of rigid PUR foams designed for district heating pipes exist, normal and microcellular foam, where the cell size for normal foam is around 0.25-0.30 mm and for microcellular foam around 0.10-0.20 mm. By reducing the cell size from 0.8 to 0.2 mm, the radiation contribution to thermal conductivity will decrease from 0.0057 to 0.0020 W·m⁻¹·K⁻¹ @ 40°C.

Conduction in cell gas mixture

The development of the production process in combination with the change of blowing agents resulted in the second and third generation of PET foams. The possibility of producing PET foam with the same type of blowing agent as for PUR seems to be solved. The cell gas content of a PET and a PUR foam is compared in Table 14.

		Cell gas concentration %-vol						
Foam	Total pressure @ 24°C (kPa)	Oxygen	Nitrogen	Carbon dioxide	Cyclo- pentane	Iso- pentane	Water vapour	Others
PUR	126	0.2	0.8	65	32	2	0.4	0.5
PET	48	2	7.4	0.4	85	0	3.9	1.5

% cyclopentane in gas phase of the total amount of cyclopentane in the cells (gas + condensed liquid) @ 24° C for the PUR foam was 100% and for the PET foam 60%.

Table 14. Cell gas content (%-vol) for a cyclopentane blown PET foam and for a PUR foam used for district heating pipes.

Total thermal conductivity

A comparison between a newly produced cyclopentane blown PET foam and a newly produced cyclopentane blown PUR foam of the same density is shown in Table 15. The PUR foam has about 10% lower thermal conductivity than the PET foam. However, the PUR foam has been developed during more than 50 years but the PET foam during less than 10 years and can thus be expected to be further developed.

Foam	Density (kg·m ⁻³)	Cell size (mm)	$\begin{matrix} \lambda_{40} \\ (W \cdot m^{\text{-1}} \cdot K^{\text{-1}}) \end{matrix}$
PUR	55	0.29-0.31	0.0255
PET	55	0.8-1.0	0.0285

Long term thermal performance

It should be noticed that the PET foam is a material under a phase of development and therefore it is not wise to draw any statement on exact figures. However, the results from the determination of the diffusion properties give a clear cut indication that the long term thermal performance of a PET foam is better then that of a PUR foam. Table 16 shows the ageing pattern of PET foam which would be 5-15 times lower than that of rigid PUR foam.

Recent studies have shown that the diffusion coefficients of oxygen and nitrogen are 2-4 times lower in rigid foam than in semiflexible PUR foam (used in flexible pipes). Therefore the aging pattern of PET insulated flexible pipes must be very favourable in comparison with PUR insulated flexible pipes.

		Ratio D _{PET} /D _{PUR}	
	20°C	40°C	60°C
			2
	P	ET foam density 144 kg·1	m ^{-s}
Nitrogen	0.06	0.05	0.04
Oxygen	0.09	0.08	0.07
Carbon dioxide	0.09	0.10	0.11
	Р	ET foam density 55 kg·n	n ⁻³
Nitrogen	0.21	0.16	0.13
Oxygen	0.25	0.22	0.20

Table 16. The ratio between the effective diffusion coefficient of PET foam and PUR foam

Table 15. Thermal conductivity (@ 40 °C (λ_{40}) of a PUR foam and a PET foam, both blown with

cyclopentane

The lower effective diffusion coefficients for PET foam will result in slower ageing which is illustrated in Figure 39. Here the calculated change of thermal conductivity (a) 10° C of the cell gas mixture over time in 35 mm thick homogeneous insulating boards stored at 23° C, 40° C and 60° C, is shown. The figure demonstrates that the decrease in insulating capacity due to the diffusion of cell gases is about ten times slower for PET foam than for PUR-foam

Figure 39. Calculated change of thermal conductivity @ 10°C of the cell gas mixture in PET and PUR boards over time for 35 mm thick carbon dioxide blown foam boards stored at different temperatures. The initial partial pressure were 80 kPa for carbon dioxide and 2,5 kPa for oxygen and nitrogen respectively.



From the determination of the diffusion coefficients of the cell gases at different temperatures in PUR and PET foam it can calculated that the activation energies are more or less the same. This means that the temperature dependencies of the thermal conductivities of the two foam types are the same.

	Activation energy E _D (J·mole ⁻¹)						
	Nitrogen	Oxygen	Carbon dioxide	Cyclopentane			
PUR ^[a]	4.5·10 ⁴	3.0·10 ⁴	$2.0 \cdot 10^4$	6.0·10 ⁴			
PET	3.5·10 ⁴	$2.5 \cdot 10^4$	$2.5 \cdot 10^4$				

^[a] M. Olsson 2001

Table 17. Activation energies for diffusion of nitrogen, oxygen, carbon dioxide and cyclopentane in PUR and PET foam.

Calculation of long term thermal performance

The long term thermal performance (LTTP) of a PET foam and a PUR foam insulated district heating pipe has been calculated by Camilla Persson (all details about the calculations are given in Appendix 1). Both pipes were assumed to have the same dimensions (DN 40/125) and the same foam density ($55 \text{ kg} \cdot \text{m}^{-3}$). The calculations were made for a service pipe temperature of 80°C and a casing temperature of 15°C during a period of 30 years. The initial cell gas contents of the PET and PUR foams were very close to those given in Table 14.

The contribution of radiation and conduction in the polymer to the thermal conductivity of insulating foams is constant over time. It is estimated at $0.011 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ @ 40°C for both foams. This is a realistic value for a PUR foam produced today but not for a PET foam since the cells a PET foam are larger (0.8-1.0mm) than the cells of a PUR foam (about 0.3mm), see Table 15. However, after further development the cell size can be expected to be the same in both foams.

The diffusion of the cell gases in the foams and in the casings over time was calculated numerically with aid of explicit finite differences. The thermal conductivity of the resulting cell gas mixture was calculated from the Wassiljeva equation with the Mason and Saxena modification.

All phases of cyclopentane (as a gas or a liquid in the cells or dissolved in the polymer matrix) were considered. The equilibrium between the partial pressure of cyclopentane in the cell gas and concentration of cyclopentane in the polymer matrix was assumed to follow Henry's law.

The radial temperature profile of the pipe is updated for each year. This has proven to give accurate results since the heat conduction process is much faster than the diffusion process and the temperature re-coupling is weak.

The result of the calculations are given in Figure 40a (PUR, 3.0mm HDPE casing), 40b (PET, 3.0mm PET casing) and 40c (PET, no casing). Due to the slow diffusion in the PET foam, the decrease of insulating capacity of the PET foam insulated pipe without casing is even less (about 6%) than that of the PUR insulated pipe with 3.0mm HDPE casing (about 16%). The PET insulated pipe with 1.0mm PET casing exhibits the slowest decrease (about 3%) of insulating capacity during 30 years among the three alternatives studied.

Note, that a lower initial thermal conductivity of the PET foam was assumed than what has been measured for PET foams produced so far and that the thermal conductivity due to radiation and conduction through the matrix was assumed equal for both foams. The calculations therefore illustrate the performance of an improved PET foam, e.g. with smaller cells.

The decrease of thermal conductivity of the PUR insulated pipe during the first two years depends upon the fact that carbon dioxide is diffuses faster out of the foam than air diffuses into the foam.

Figure 40 a. Thermal conductivity over time for a PUR insulated district heating pipe (DN40/125). Service pipe temperature 80 °C and casing temperature 15 °C. Casing material HDPE, thickness 3.0 mm.

Figure 40b. Thermal conductivity over time for a PET insulated district heating pipe (DN40/125). Service pipe temperature 80 °C and casing temperature 15 °C. Casing material PET, thickness 3.0 mm.

The insulation has an improved quality of PET foam, e.g. with smaller cells than what is possible to produce today.

Figure 40c. Thermal conductivity over time for a PET insulated district heating pipe (DN40/125). Service pipe temperature 80 °C and outer surface temperature 15 °C. The pipe has no casing.

The insulation has an improved quality of PET foam, e.g. with smaller cells than what is possible to produce today.






3.3 Environmental performance

From the present study it can be concluded that PET foam has the potential to compete successfully in terms of environmental performance with cyclopentane blown PUR foam as insulating foam for district heating pipes. After the end of this environmental study, B.C.Foam succeeded in producing cyclopentane blown PET foam of low density. However, commercial methods to produce PET foam for the insulation of district heating pipes must be developed. Utilization of recycled PET can reduce the environmental impacts from the production phase of the pipes life cycle and would contribute to the efficient use of resources in society.





Figure 40. Characterised environmental impacts from the three life cycle phases for the studied impact categories:

a) global warming potential,

b) acidification potential and

c) resource depletion.

All values are given for 1 m DN100 pipe network (1m flow pipe and 1 m return pipe) over 30 years of use. Heat losses described by environmental impacts from average Swedish district heating fuel mix.

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Appendix 1

Comparison of the long-term thermal performance of a polyethylene terephthalate (PET) foam and a polyurethane (PUR) foam insulated district heating pipe

Camilla Persson, Department of Civil and Environmental Engineering, Chalmers University of Technology, Sweden

The long-term thermal performance (LTTP) of a PET foam and a PUR foam insulated district heating pipe has been calculated. Both pipes were assumed to have the same dimensions (DN40/125) and the same foam density (55 kg·m⁻³). The PUR foam insulated pipe had a 3.0 mm casing of high-density polyethylene (HDPE). Three casing alternatives were considered for the PET foam insulated pipe: a 1.0 mm and a 3.0 mm thick casing of PET, as well as no casing at all. The calculations were made for a service pipe temperature of 80°C and a casing temperature of 15°C over a period of 30 years.

The cell gas compositions of the foams change over time due to gas diffusion and impacts on the thermal conductivity and heat flow from the pipes. The LTTP-calculations were performed according to a method presented in [1, 2], briefly described below under Model.

Model

Transport of each gas through the foams was assumed to satisfy the radial diffusion equation A1. The diffusion equation states that the change of the total concentration of the gas in the foam c_{tot} (mol·m⁻³) over time t (s) is given by the net inflow, expressed as the space derivative of the molar mass flux j (mol·m⁻²·s⁻¹).

$$\frac{\partial c_{tot}}{\partial t} = -\frac{1}{r} \cdot \frac{\partial}{\partial r} (r \cdot j)$$
(A1)

The mass flow equals the temperature dependent diffusion coefficient δ (m²·s⁻¹) times the concentration derivative (eq A2).

$$j = -\delta_c \cdot \frac{\partial c}{\partial r}$$
(A2)

The total concentration of the gas is all phases' contributions summed (eq A3); the concentration in the cell gas c (mol·m⁻³ gas), the concentration dissolved in the polymer matrix c_{pol} (mol·m⁻³ matrix) and the condensed liquid concentration c_{liq} (mol·m⁻³). The gas fraction of the foam is f_g (-).

$$\mathbf{c}_{\text{tot}} = \mathbf{c}_{\text{pol}}(1 - \mathbf{f}_{g}) + \mathbf{c} \cdot \mathbf{f}_{g} + \mathbf{c}_{\text{liq}} = \left[\mathbf{S} \cdot \mathbf{R} \cdot \mathbf{T} \cdot (1 - \mathbf{f}_{g}) + \mathbf{f}_{g}\right] \cdot \mathbf{c} + \mathbf{c}_{\text{liq}}$$
(A3)

The equilibrium between the partial pressure of cyclopentane in the cell gas p (Pa) and the concentration of cyclopentane in the polymer matrix c_{pol} (mol·m⁻³) was assumed given by the solubility coefficient S (mol·m⁻³·Pa⁻¹) according to Henry's law (eq A4),

$$\mathbf{c}_{\mathrm{pol}} = \mathbf{S} \cdot \mathbf{p} \tag{A4}$$

Equation A4 and the ideal gas law (molar gas constant $R=8.314 \text{ J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ were used to rewrite the concentration of cyclopentane dissolved in the polymer in equation A3 into an expression for the gas phase concentration. The concentration of condensed liquid cyclopentane was determined so that the saturation pressure of cyclopentane was not exceeded.

For oxygen, nitrogen and carbon dioxide only the gas phase was considered. The solubility of oxygen, nitrogen and carbon dioxide in the polymer matrix was assumed to be low. The solubility coefficient of oxygen in PET is about $6\cdot10^{-5}$ mol·m⁻³·Pa⁻¹ at 25°C

[3] and the solubility coefficient of nitrogen in PET is around $2 \cdot 10^{-5}$ mol·m⁻³·Pa⁻¹ at 50°C [4]. The value of these coefficients should be compared to that of cyclopentane of around $2 \cdot 10^{-3}$ mol·m⁻³·Pa⁻¹ at 25°C [5].

The steel service pipes were considered gas tight, while the casings were considered to act as diffusion barriers. The outer radius of the service pipe is $r_f(m)$. The inner radius of the casing is $r_{ins}(m)$ and the outer radius $r_c(m)$.

$$\mathbf{j}\big|_{\mathbf{r}=\mathbf{r}_{\mathbf{f}}} = \mathbf{0} \tag{A5}$$

$$2 \cdot \pi \cdot \mathbf{r}_{\text{ins}} \cdot \mathbf{j}\Big|_{\mathbf{r}=\mathbf{r}_{\text{ins}}} = \frac{\mathbf{c}(\mathbf{r}_{\text{ins}}) - \mathbf{c}_{\text{e}}}{Z_{\text{c}}} \qquad Z_{\text{c}} = \frac{\ln(\mathbf{r}_{\text{c}}/\mathbf{r}_{\text{ins}})}{2 \cdot \pi \cdot \mathbf{P} \cdot \mathbf{R} \cdot \mathbf{T}_{\text{c}}}$$
(A6)

The total mass flow through the casing is given by the concentration difference between the inside $c(r_{ins})$ and the outside c_e , divided by the resistance of the casing $Z_c (s \cdot m^{-2})$. The permeability coefficient of the casing is P (mol·m⁻¹·s⁻¹·Pa⁻¹) and the casing temperature is T_c (K).

The radial heat flow q (J·s⁻¹·m⁻²) through the insulation is given by the thermal conductivity λ (W·m⁻¹·K⁻¹) times the temperature derivate. The thermal conductivity of the foam depends on the cell gas composition.

$$\mathbf{q} = -\lambda \cdot \frac{\partial \mathbf{T}}{\partial \mathbf{r}} \tag{A7}$$

The diffusion problem was solved numerically over time with explicit finite differences. A steady-state temperature profile through the foam was used. It was updated each year, to account for the change of the cell gas composition. Constant temperatures of the steel service pipes $(80^{\circ}C)$ and the casings $(15^{\circ}C)$ were assumed.

Initial cell gas content

The initial cell gas content differs between the PUR and the PET foam (Table 1). The values were based on the experience from earlier determinations made at Chalmers [6, 7, 8] and are very close to those given in Table 14 (see Chapter 3.2).

	Cell gas content (vol-%)		
	PUR-foam	PET-foam	
Oxygen	0.2	2.4	
Nitrogen	0.8	9.6	
Carbon dioxide	67	0	
Cyclopentane	32	88	
% Cyclopentane in gas phase (the rest in liquid phase) @24°C	100	60	
Total pressure (kPa)	126	48	

Partial pressures in ambient air: CO2 0 kPa, N2 78 kPa, O2 21 kPa

Table 1. Initial cell gas content of the PUR and PET foam at 25°C

Diffusion through the foams

The diffusion coefficient for each gas in each foam δ (m²·s⁻¹) was calculated from equation A8.

$$\delta = \mathbf{D}_{0} \cdot \exp\left[\frac{-\mathbf{E}_{\mathrm{D}}}{\mathbf{R} \cdot \mathbf{T}}\right] \cdot \left(\mathbf{S} \cdot \mathbf{R} \cdot \mathbf{T} \cdot (1 - \mathbf{f}_{\mathrm{g}}) + \mathbf{f}_{\mathrm{g}}\right)$$
(A8)

The diffusion parameters are the pre-exponential factor D_0 (m²·s⁻¹) and the activation energy for diffusion E_D (J·mol⁻¹). These were determined to fit the effective diffusion coefficients at different temperatures given in literature. The effective diffusion coefficients had been determined according to a method described in [9]. Foam samples (cylinders or boards) were stored at isothermal conditions. The mean partial pressures in the samples were measured after different times of storage. The effective diffusion coefficients were determined by matching the calculated changes of the mean partial pressures according to equation A9, to the measured values. At constant temperature, the partial pressure p is interchangeable with the concentration c in equation A9 for ideal gases.

$$\frac{\partial \mathbf{p}}{\partial t} = -\mathbf{D}_{\rm eff} \cdot \frac{\partial^2 \mathbf{p}}{\partial x^2} \tag{A9}$$

Assuming instant equilibrium between the gas and dissolved phases radial diffusion is described by equation A1. If the effective diffusion coefficients were determined for foams without condensed liquid cyclopentane, the diffusion coefficient δ can be determined based on the diffusion parameters by equation A8.

The diffusion parameters for the PUR foam were derived from [6,7]. For the PET foam the values are from [8]. The diffusion parameters for cyclopentane in PET were decided upon to get approximately the same relation between diffusion in PET and PUR as for the other gases. With the chosen values $D_0 \exp(-E_D/(R \cdot T))$ for cyclopentane in PET equals 1/6 of that for cyclopentane in PUR.

Diffusion parameters				
	PUR foam PUR foam PET foam		PET foam	
	D_0	Ed	D_0	Ed
	$m^2 \cdot s^{-1}$	J•mol ⁻¹	$m^2 \cdot s^{-1}$	J·mol ⁻¹
Oxygen	3.39·10 ⁻⁴	$4.16 \cdot 10^4$	1.13·10 ⁻⁷	$2.51 \cdot 10^4$
Nitrogen	$1.02 \cdot 10^{-2}$	$5.42 \cdot 10^4$	9.08·10 ⁻⁷	$3.50 \cdot 10^4$
Carbon dioxide	$4.77 \cdot 10^{-4}$	$3.98 \cdot 10^4$	-	-
Cyclopentane	1.69.10-7	$3.52 \cdot 10^4$	$2.82 \cdot 10^{-8}$	$3.52 \cdot 10^4$

Permeability of the casings

Arrhenius relationships were assumed for the permeability coefficients of the casings P $(mol \cdot m^{-1} \cdot s^{-1} \cdot Pa^{-1})$ according to equation A10.

$$\mathbf{P} = \mathbf{P}_0 \cdot \exp\left[\frac{-\mathbf{E}_p}{\mathbf{R} \cdot \mathbf{T}}\right] \tag{A10}$$

The permeability parameters of the polyethylene casing material were derived from values given in [6] for oxygen, nitrogen and carbon dioxide and from [6,10] for cyclopentane. The values calculated at 15°C according to these expressions were used.

Table 2. Diffusion parameters for PUR and PET foam used in the calculations

Table 3. Permeability parameters for the HDPE casing

Table 4. Permeabilitycoefficients for solid PET at

25 and 15 $^{\circ}C$

	Permeability of HDPE casing		
	P ₀	Ep	P @15 °C
	mol·m ⁻¹ ·s ⁻¹ ·Pa ⁻¹	J-mol ⁻¹	mol·m ⁻¹ ·s ⁻¹ ·Pa ⁻¹
Oxygen	$2.97 \cdot 10^{-10}$	$3.48 \cdot 10^4$	$1.44 \cdot 10^{-16}$
Nitrogen	$5.44 \cdot 10^{-10}$	$3.92 \cdot 10^4$	$4.28 \cdot 10^{-17}$
Carbon dioxide	5.58·10 ⁻¹¹	$2.70 \cdot 10^4$	$7.21 \cdot 10^{-16}$
Cyclopentane	$1.31 \cdot 10^{-13}$	$9.94 \cdot 10^{3}$	$2.06 \cdot 10^{-15}$

Permeabilities of oxygen and nitrogen in PET at 25 °C from [11] have been recalculated to 15° C using an activation energy $E_P = 33600 \text{ J} \cdot \text{mol}^{-1}$ from [3]. The activation energy was calculated based on values given for oxygen but was assumed valid for all gases. For cyclopentane the PET casing was assumed to have no resistance.

	$\begin{array}{c} P @25 \ ^{\circ}C \\ mol \cdot m^{-1} \cdot s^{-1} \cdot Pa^{-1} \end{array}$	$\begin{array}{c} P @ 15 ^{\circ}C \\ mol \cdot m^{-1} \cdot s^{-1} \cdot Pa^{-1} \end{array}$
Oxygen	$1.34 \cdot 10^{-17}$	8.36·10 ⁻¹⁸
Nitrogen	$1.79 \cdot 10^{-18}$	$1.11 \cdot 10^{-18}$

Solubility in the foam matrix

The temperature dependence of the solubility coefficients S (mol·m⁻³·Pa⁻¹) was assumed to follow an Arrhenius relationship. The solubility parameters in the PUR matrix were from [12]. In [5] the solubility coefficient of cyclopentane at 23 °C in PET was determined to $2.4 \cdot 10^{-3}$ mol·m⁻³·Pa⁻¹. Due to lack of information, the same activation energy was assumed for both matrixes.

$$\mathbf{S} = \mathbf{S}_0 \cdot \exp\left[\frac{-\mathbf{E}_s}{\mathbf{R} \cdot \mathbf{T}}\right] \tag{A11}$$

	PUR foam		PET foam	
	S ₀	Es	S ₀	Es
	mol·m ⁻³ ·Pa ⁻¹	J•mol ⁻¹	mol·m ⁻³ ·Pa ⁻¹	J∙mol ⁻¹
Cyclopentane	$1.3 \cdot 10^{-5}$	$-16 \cdot 10^3$	$3.6 \cdot 10^{-6}$	$-16 \cdot 10^3$

Thermal conductivity

The heat transport through the foam takes place by conduction through the gas and the matrix as well as radiation. The thermal conductivity of the foams was modelled as the sum of the different transports' contributions.

The contribution of radiation and conduction in the polymer was assumed constant over time. It was estimated at 0.011 W·m⁻¹·K⁻¹ at 40°C for both foams. This is a value that is approximately valid for a PUR foam produced today. However, the PET must be further developed in order to lower its thermal conductivity. Cells of the same small size as in the PUR foam may decrease the thermal conductivity of the foam.

The thermal conductivity of the gas in the cells was calculated according to Wassiljeva's equation with the Mason and Saxena modification that uses critical temperatures and pressures [13]. The critical temperatures and pressures for oxygen, nitrogen and carbon dioxide were taken from [14]. Data for cyclopentane was from [15,16]. The temperature dependencies of the thermal conductivities of the gases were from [17].

Table 5. Solubility of cyclopentane in the PUR- and the PET-matrix.

Results

0.032

0



Thermal conductivity [W/(m·K)] 0.03 0.028 0.026 0 5 10 15 20 25 Time [years] 40 Gas phase concentration [mol/m3] cycyclopentane 30 carbon dioxide oxygen nitrogen 20 10

5

10

15

Time [years]

20

25

30

Figure 1. Thermal conductivity over time for a PUR foam insulated district heating pipe (DN40/125).

Service pipe temperature $80^{\circ}C$ and casing temperature $15^{\circ}C$.

Casing material HDPE, thickness 3.0 mm.

Figure 2.The average cell gas concentrations over time for a PUR foam insulated district heating pipe (DN40/125).

Service pipe temperature 80°*C and casing temperature* 15°*C*.

Casing material HDPE, thickness 3.0 mm.



30

Figure 3.Radial concentration profiles of cyclopentane in a PUR foam insulated district heating pipe (DN40/125) at different times of ageing (0, 10, 20 and 30 years).

Service pipe temperature 80 °C and casing temperature 15 °C.

Casing material HDPE, thickness 3.0 mm.

All the different phases of cyclopentane – gas, condensed liquid and dissolved in the polymer matrix – have been considered.



Radial position in the insulation [m]

PET-foam insulated pipe with 1.0 mm thick PET-casing

Figure 4. Thermal conductivity over time for a PET foam insulated district heating pipe (DN40/125).

Service pipe temperature $80^{\circ}C$ and casing temperature $15^{\circ}C$.

Casing material PET, thickness 1.0 mm.

The insulation has an improved quality of PET foam, e.g. with smaller cells than what is possible to produce today.

Figure 5. The average cell gas concentrations over time for a PET foam insulated district heating pipe (DN40/125).

Service pipe temperature $80^{\circ}C$ and casing temperature $15^{\circ}C$.

Casing material PET, thickness 1.0 mm.





Figure 6. Radial concentration profiles of cyclopentane in a PET foam insulated district heating pipe (DN40/125) at different times of ageing (0, 10, 20 and 30 years).

Service pipe temperature 80°C and casing temperature 15°C.

Casing material PET, thickness 1.0 mm.

All the different phases of cyclopentane – gas, condensed liquid and dissolved in the polymer matrix – have been considered.



Radial position in the insulation [m]

PET-foam insulated pipe with 3.0 mm thick PET-casing

Figure 7. Thermal conductivity over time for a PET foam insulated district heating pipe (DN40/125).

Service pipe temperature $80^{\circ}C$ and casing temperature $15^{\circ}C$.

Casing material PET, thickness 3.0 mm.

The insulation has an improved quality of PET foam, e.g. with smaller cells than what is possible to produce today.

Figure 8. The average cell gas concentrations over time for a PET foam insulated district heating pipe (DN40/125).

Service pipe temperature 80°*C and casing temperature* 15°*C*.

Casing material PET, thickness 3.0 mm.





Figure 9. Radial concentration profiles of cyclopentane in a PET foam insulated district heating pipe (DN40/125) at different times of ageing (0, 10, 20 and 30 years).

Service pipe temperature 80°C and casing temperature 15°C.

Casing material PET, thickness 3.0 mm.

All the different phases of cyclopentane – gas, condensed liquid and dissolved in the polymer matrix – have been considered.



Radial position in the insulation [m]

PET-foam insulated pipe without any casing

Figure 10. Thermal conductivity over time for a PET foam insulated district heating pipe (DN40/125).

Service pipe temperature 80°C and outer surface temperature 15°C.

The pipe has no casing.

The insulation has an improved quality of PET foam, e.g. with smaller cells than what is possible to produce today.

Figure 11. The average cell gas concentrations over time for a PET foam insulated district heating pipe (DN40/125).

Service pipe temperature 80°C and outer surface temperature 15°C.

The pipe has no casing.



Figure 12. Radial concentration profiles of cyclopentane in a PET foam insulated district heating pipe (DN40/125) at different times of ageing (0, 10, 20 and 30 years).

Service pipe temperature 80°C and outer surface temperature 15°C.

The pipe has no casing.

All the different phases of cyclopentane – gas, condensed liquid and dissolved in the polymer matrix – have been considered.



Radial position in the insulation [m]

Comparison of a PUR foam and a PET foam insulated pipe

In Table 6 the calculated equivalent thermal conductivities of the insulations λ_{eq} (W·m⁻¹·K⁻¹) and mean heat flows from the pipes Q_{eq} (W·m⁻¹) over 30 years of use are stated. The equivalent thermal conductivity of the insulation is the constant thermal conductivity of the whole cross-section over the time period that yields the same heat loss as the actual time and space dependent thermal conductivity. In the same way, the mean heat flow equals the constant heat flow over the time period that yields the same total heat loss as the actual time dependent heat flow.

Note, that a lower initial thermal conductivity of the PET foam was assumed than what has been measured for PET foams produced so far [8]. The thermal conductivity due to radiation and conduction through the matrix was assumed equal for both foams. The calculations therefore illustrate the performance of an improved PET foam, e.g. with smaller cells.

Туре	of pipe	Equivalent over	a 30 years	
Insulation	Casing	Thermal conductivity of	Heat flow Q_{eq} (W·m ⁻¹)	
	_	the foam λ_{eq} (W·m ⁻¹ ·K ⁻¹)	1	
PUR	HDPE 3 mm	0.0294	13.3	
PET	PET 3 mm	0.0256	11.6	
PET	PET 1 mm	0.0261	11.8	
PET	No casing	0.0288	13.1	

The insulation thickness of all pipes is the same. This means that the pipe with 1 mm casing has a 4 mm smaller outside diameter than the other pipes with casings.

Table 6.The equivalent thermal conductivity and heat losses for PUR and PET insulated district heating pipes (DN40/125) over an operation period of 30 years.

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