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Selection of low GWP participating gas for a passive cooling skylight

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Abstract

Rising temperatures as a result of climate make the cooling of building envelopes and creating thermal comfort for more and more people a challenge while energy use must become more efficient. In addition to active (electricity-driven) systems, passive cooling methods are being developed for night-time as well as day-time cooling of buildings. A passive cooling skylight under development at Åbo Akademi has demonstrated a night-time passive cooling effect of ~ 100 W/m². Its performance depends strongly on the gas used inside the skylight, that will pick up thermal radiation (at long wavelengths, LW) via a lower window and after a natural convection transfer inside the skylight, release the heat to the sky via an upper window. Proof-of-concept work at ÅA involved comparing the use of air with the performance when using carbon dioxide CO₂, ammonia NH₃ or pentafluoro ethane, i.e. hydrofluorocarbon refrigerant HFC-125. Best results reported were obtained with the latter, but as a result of the 2016 Kigali Amendment to the 1986 Montreal Protocol on HFCs, an alternative for HFC-125 needs to be found, because of its global warming potential (GWP). With GWP = 1 for CO₂, = 0 for NH₃, for HFC-125 the GWP = 3500. Since a passive cooling skylight can be ranked as a refrigeration installation it cannot contain a gas with a GWP >150 after 2030 under European regulation. The prime material property of a suitable gas is a high emissivity / absorption in the LW range 8-14 μ m, also known as the atmospheric window, where, apart from water vapour, nothing prevents radiative heat transfer from Earth directly to space. The hypothesis was that HFC-152a or HFC-41, with GWP values < 150 could replace HFC-125 for this skylight application. Besides GWP, other features that must not prevent the widespread use of the technology are flammability and chemical / physical stability in general, toxicity and, of course, costs. CFD simulations (Ansys Fluent) were used to calculate the passive cooling (LW) heat fluxes, temperatures, the convection flow field, and the transported heat inside the skylight, comparing the mentioned gases. The results show that the performance of HFC-152a (117.8 W/m²) and slightly less so HFC-41 (115.4 W/m²), both with a GWP < 150, can match the performance achieved so far with HFC-125 (117.3 W/m²), compared to 100.5 W/m² with air.

1 Introduction

There is general consensus especially in the scientific community that climate change including global warming is caused primarily by combustion of fossil fuels, generating CO₂, CH₄ and N₂O emissions, and that extreme temperature weather events will become more frequent. One result of this is a refugee crisis caused by more than wars alone, while another outcome is R&D followed by market-scale use of passive cooling systems that, contrary to active cooling systems do not use an external energy input like electricity [1]. The use of shades, smart orientation of buildings, façades and especially windows with respect to the sun, cold ponds or water sprays, storages of snow and ice and simple mechanical devices were the most important ways of keeping temperatures inside buildings low until the 19th century. Absorption cooling and electricity-driven heating, ventilation and air conditioning (HVAC) have been developed since then, and nowadays the vapour-compression refrigeration cycles is the most widely used, electricity-driven HVAC system, often integrated with desiccant processes for humidity control [2,3].

Passive cooling systems may comprise surfaces that not only reflect solar irradiation but also emit longer wavelengths heat radiation, for example so-called cool roofs, and windows including skylights, in both cases benefitting from modern materials that include nano-particles (NPs) [4-6]. In this paper, the development and testing of a transmissive skylight is reported, with a design that makes it stand out from other passive cooling (skylight) processes. The novelty lies with the use of NPs to control the influx of solar irradiation into a building envelope combined with the use of a participating (“greenhouse”) gas inside the skylight that absorbs and later emits the heat. The selection of a suitable gas for this is the subject of this paper.

The transmissive passive cooling skylight under development at Åbo Akademi (ÅA) has demonstrated a night-time passive cooling effect of around 100 W/m². Its performance depends strongly on the gas used inside the skylight: that gas will pick up thermal radiation at long wavelengths (LW, say 5 – 20 μm) via a lower window and after a natural convective heat transfer inside the skylight, bring the heat in (visual) contact with the sky via an upper window. Behind the sky, the universe presents a vast 3 K heat sink. Cleartran® ZnS window glass (CWG) used offers the necessary LW transmittance for taking in heat from a building space into the skylight and later transporting it to the sky.

Proof-of-concept work at ÅA – see Figure 1 - involved comparing the use of air with the performance obtained when using carbon dioxide CO₂, ammonia NH₃ or hydrofluorocarbon refrigerant HFC-125/R-125 = pentafluoro ethane, as reported in e.g. [7-10] and a PhD thesis [11]. Best results reported were obtained at ÅA with the HFC-125, but as a result of the 2016 Kigali Amendment to the 1986 Montreal Protocol on the production and use of HFCs, an alternative for HFC-125 needs to be found (at least for application in the EU/EEA area), because of the global warming potential (GWP) [12].

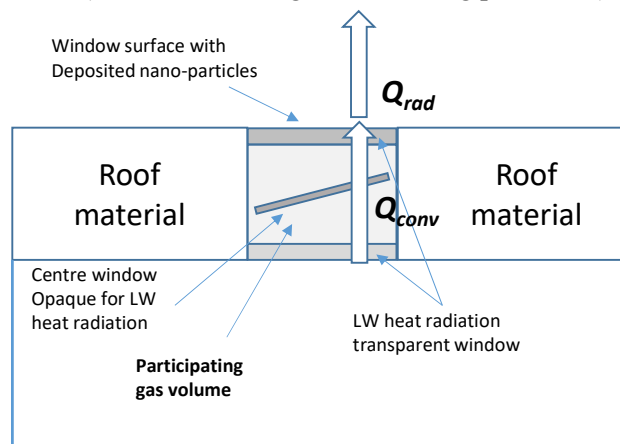


Figure 1: Schematic of the ÅA transmissive skylight (typical dimensions 50 cm length x 50 cm width x 10 cm height) *Note* that for the current design the centre window is removed; the convection inside the skylight then arises from asymmetric positioning of LW transparent sections in “standard” (opaque for LW radiation) glass.

While GWP = 1 for CO₂ and GWP = 0 for NH₃, for HFC-125 the GWP = 3500. Since a passive cooling skylight can be ranked as a refrigeration installation it cannot contain a gas with a GWP >150 after year 2030 under European regulation [13]. More recent reporting on European Commission proposals for air conditioners and heat pumps (AC-HP) gives GWP < 150 until 2027, 2029 or 2033 depending on system type and size, and GWP < 750 for systems > 12 kW, until GWP < 150 in 2033 also for these [14]. This excludes not only HFC-125 but also, for example, the currently popular difluoro methane, HFC-32/R-32 with a GWP of 675 is removed from the list of potential alternatives for HFC-125 for the passive cooling skylight.

The prime material property of a suitable gas is a high emissivity / absorption in the LW range 8-14 μm, also known as the “atmospheric window” where, apart from water vapour, nothing prevents radiative heat transfer from an object on Earth directly to space [15]. This seems to contradict the GWP < 150 requirement, but the specific wavelengths requirements define what is needed versus will be allowed.

This paper reports on the search for and identification of a suitable gas for the ÅÅ passive cooling skylight – see Figure 1 - with high emissivity /absorption for LW thermal radiation in the atmospheric window range. Besides this requirement, other features that must not prevent the widespread use of the technology are flammability and chemical / physical stability in general, toxicity and, of course, costs. This is part of developments of making the ÅÅ skylight effective also during daytime; see our recent work that addresses the use of nano-particulate material on the outer window in order to control solar irradiation entering the building space [16,17] and the asymmetric positioning of window sections transparent for LW thermal radiation in standard window glass (SWG) [18] as also used below.

2 GWP of gases suitable for a passive cooling skylight

2.1 Thermal radiation properties

For a gas to be suitable for a transmissive passive cooling skylight, transporting LW thermal radiation out of a building space, a high emissivity, ϵ , and absorptivity, α , is needed. Then heat can be emitted to the (upper) atmosphere and eventually, with a clear sky, the cold universe behind it via the 8-14 μm atmospheric window. For the modest (< 50 K) temperature differences considered here, Kirchoff’s law is used that equates emissivity to absorptivity, $\epsilon = \alpha$, for the four wavelength bands use here: short wavelength (SW) < 4 μm) and three long wavelength (LW) ranges 4-8 μm, 8-14 μm and > 14 μm, respectively. (See [19] for the development of the four-wavelength band concept used here.) For the window glass, LW transmitting sections of Cleartran ® ZnS (CWG) are used, while as ongoing parallel work the use of dispersed nanoparticles on the outside window is being optimised for selectively blocking out solar (SW) irradiation.

Table 1 lists properties of gases that have been used (air, CO₂, NH₃, HFC-125) or would be potentially suitable for the ÅÅ skylight, including their GWP. For the air, CO₂, and NH₃ the emissivity (assumed = absorptivity) for the four wavelength bands could be calculated using [20-23]). Assuming reflectance $\rho = 0$, this implies transmittance

$$\tau = 1 - \alpha = 1 - \epsilon \quad (1)$$

For HFC-125, HFC-152a and HFC-41 emissivity data is calculated using NIST “webbook” data [24-26], unfortunately data for HFC-32 was found in a format that made it unsuitable for use here [27,28] so that gas had to be excluded at this point.

The NIST data sets are mostly reported as absorbance, A (-), which is related to transmittance τ (-) as

$$A = -\log(\tau) \quad (2)$$

For the calculations using Ansys Fluent both the emissivity ϵ (= absorptivity α) and the absorption coefficient κ (m⁻¹) must be given, which for an optical path L (m) is found from Lambert-Beer’s equation

$$\tau = \frac{\text{radiation passing}}{\text{radiation entering}} = 1 - \alpha = 1 - \varepsilon = e^{-\kappa L} \quad (3)$$

$$\rightarrow \kappa = \frac{-\ln(\tau)}{L} = \frac{-2.303 \cdot 10 \log(\tau)}{L} = \frac{-2.303 \cdot A}{L} \quad (4)$$

for a gas with reflectivity = 0 (-) and refractive index = 1 (-), and $2.303 \approx \ln(10)$. Often (which gives more detail about the experimental procedure) the absorbance A is reported as $A = \alpha' \cdot c \cdot L$ with a molar absorptivity α' (mol/(mol·m)), concentration c of the active species (mol/mol), and path L (m).

For the CFD model simulations given below, it was necessary to correct for the pathlength L versus the pathlength used for determining emissivities. For air, CO₂ and NH₃, the literature models [20-23] allowed calculations for any path length, temperature, and pressure, while the NIST database reports for 296 K, 101.3 kPa and L = 20 cm for the Bruker IFS66V analyzer used. The skylight model used for the simulations given in this paper would, for the geometry used and boundary conditions specified, require input data for 101.3 kPa, 288 K and L = 30 cm for which a re-calculation from 296 K, 101.3 kPa and L = 20 cm data is needed.

Using the detailed models for air, CO₂ and NH₃, it could be determined that the values found for emissivities at 101.3 kPa, 288 K and L = 30 cm are 10% higher than the values at 101.3 kPa, 296 K and L = 20 cm. Thus, in Table 1, values for air, CO₂ and NH₃ are for both sets calculated using the detailed literature models, while for HCF-125, HFC-152a and HFC-41 the values for 101.3 kPa, 288 K and L = 30 cm are taken to be 110% × the values found from the NIST data, measured for 101.3 kPa, 296 K and L = 20 cm.

Table 1: Gases proven or potentially suitable for a transmissive passive cooling skylight. The absorbance (= emissivity) for the 8-14 μm range given in underlined

Gas	Absorbing / emitting wavelengths (μm)	Emissivity (-) (101.3 kPa, 296 K, 20 cm)	Emissivity (-) (101.3 kPa, 288 K, 30 cm)	GWP 100 years (-)	Atmospheric lifetime (years)
Air	Weak H ₂ O, CO ₂ , CH ₄	SW < 4 μm 8·10 ⁻⁶ LW 4-8 μm 0.0019 LW 8-14 μm <u>0.0003</u> LW > 14 μm 0.1084	6·10 ⁻⁶ 0.0016 <u>0.0004</u> 0.1186		
CO ₂	2.7; 4.3; 9.4; 10.4; 15	SW < 4 μm 1·10 ⁻⁵ LW 4-8 μm 0.0020 LW 8-14 μm <u>0.0005</u> LW > 14 μm 0.1231	7·10 ⁻⁶ 0.0017 <u>0.0006</u> 0.1354	1	100 (GWP=1)
NH ₃	2.9; 3.0; 6.15; 10.5	SW < 4 μm 0.0259 LW 4-8 μm 0.0864 LW 8-14 μm <u>0.4713</u> LW > 14 μm < 10 ⁻⁶	0.0292 0.0973 <u>0.5310</u> < 10 ⁻⁶	0	Not relevant
HFC-125 Penta fluoro ethane	7.7; 8.3; 8.8; 11.6; 13.8; 17.7	SW < 4 μm 0.0927 LW 4-8 μm 0.1640 LW 8-14 μm <u>0.3064</u> LW > 14 μm 0.2130	0.1019 0.1804 <u>0.3371</u> 0.2343	3500 [29] 3450 [30]	29 [29] 30 [30]
HFC-152a 1,1- difluoro ethane	3.3; 7.1; 8.8; 10.8	SW < 4 μm 0.0140 LW 4-8 μm 0.0214 LW 8-14 μm <u>0.1459</u> LW > 14 μm 0.0249	0.0154 0.0236 <u>0.1604</u> 0.0273	124 [29] 148 [30]	1.4 [29] 1.6 [30]
HFC-41 Fluoro methane	3.4; 6.9; 9.5	SW < 4 μm 0.2771 LW 4-8 μm 0.1198 LW 8-14 μm <u>0.3473</u> LW > 14 μm 0.04994	0.3048 0.1317 <u>0.3812</u> 0.0549	116 [30]	2.8 [30]

The data in Table 1 suggest that HFC-152a and HFC-41 would be good, GWP < 150, alternatives for (somewhat nasty to work with) ammonia, NH₃. Still, for HFC-41 and HFC-152a the handling may require support from certified specialists which complicates R&D activities like what is reported here.

2.2 Flammability and toxicity

Another property important from a health and safety point of view is flammability, ranked 1 (no flame propagation), 2L (lower burning velocity), 2 (lower flammability) or 3 (higher flammability), combined with toxicity levels A (low) or B (high), under ISO standard 817:814 (2014, reviewed/confirmed 2020). With ranking A1 for CO₂, and B2L for NH₃, for HFC-41 the ranking is A2L, and for HFC-152a it is A2.

3 Passive cooling heat transfer simulations

Using the emissivity data for four wavelength bands given in Table 1, CFD model simulations using Ansys Fluent 2024 were made to calculate the passive cooling heat transfer rate. Having done experimental work using air, CO₂ (R-744), NH₃ (R-717) and HFC-125 as the participating gas inside the skylight [8, 10], the aim is to identify a gas that can replace HFC-125 but also NH₃. On the other hand, if any of the gases HFC-41 or HFC-152a, performs similar to NH₃, and we cannot acquire permission for using HFC-41 or HFC-152a in our laboratory, then at least our R&D can continue with NH₃ while eventually for a commercial consumer products a gas like HFC-41 or HFC-152a may be used.

3.1 The Fluent Ansys model

For the radiative heat transfer simulations (using Ansys Fluent 2024 R1), a skylight with dimensions length × width × height = 45 cm × 30 cm × 20 cm is considered – see the figures given below – with 10 cm length x 30 cm width CWG sections that are transparent ($\tau \approx 0.75$) for LW radiation. This shape gives path $L = (\text{length} \times \text{width} \times \text{height})^{1/3} = 0.3$ m.

Recently it was determined that for the SW wavelength region the transmittance of Cleartran ® ZnS (CWG) ≈ 0.75 as well, while the reflectance $\rho \approx 0.25$ [17]. This shows that the absorptivity = emissivity $\alpha = \varepsilon$ for the SW range must be ≈ 0 , which presumably also applies to the LW regions. The remainder of the top and bottom windows are taken to be standard window glass which is transparent $\tau \approx 0.1$ for SW radiation but opaque for LW radiation. The side walls are taken to be a material like wood, non-conducting but with emissivity $\varepsilon = 0.85$ for SW and LW radiation.

The simulations were made using a 3 mm HEX mesh with approx. 10^6 nodes and elements, for 10000 calculation steps. For the natural convection flow simulation, the viscous-laminar model was used; for the thermal radiation the discrete ordinates (DO) model is used with extended gray band model for four wavelength bands (SW + 3×LW: 0 - 4 μm + 4 - 8, 8 - 14 and 14 - 50 μm , respectively).

As for boundary conditions: for the space (a building or room) below the skylight a temperature $T = 298$ K is used, for the ground level surroundings above the skylight a temperature $T = 278$ K and for the sky a temperature $T = 268$ K is taken. (The values at 288 K for emissivities of the gases inside the skylight follows as the average of 278K and 298 K.) Modest heat convection is quantified via heat transfer coefficients taken to be $5 \text{ W/m}^2 \cdot \text{K}$ for below and above the skylight.

3.2 Simulation results – heat flows and temperatures

Table 2 lists temperatures and heat flows through the skylight using the different gases inside the skylight. Thermal radiation heat flows leaving the skylight range up to 17% higher with a participating gas compared to the 100 W/m^2 obtained with air. Interestingly, the performance of HFC-125 may be achievable with low GWP gas HFC-152a as well.

Temperature values given are averages for the outside surface of that material. Average velocities for the natural convection inside the skylight are of the order of 5 – 7 cm/s, similar to what was found earlier [20] for air and CO₂. The volumetric absorbed and emitted radiation values are slightly different as a

result of the density of the gas absorbing (just above the lower CWG window) being lower than that of the gas emitting (from just below the upper CWG window section). Interestingly, the high value for this found for HFC-125, most certainly as a result of its high emissivity does not give more passive cooling.

Table 2: Simulation results: heat flows, temperatures, convection velocities and transported radiation heat. CWG = Cleartran ® ZnS glass, SWG = standard window glass

Gas	Passive cooling heat flow (8-14 μm) (W)	Temperature CWG window top / bottom (°C)	Temperature SWG window top / bottom (°C)	Convection flow average velocity (m/s)	Volumetric absorbed / emitted radiation (W/ m ³)
Air	100.5	274.3 / 292.1	282.9 / 298.0	0.065	826 / 811
CO ₂	101.9	274.1 / 292.5	282.8 / 298.0	0.062	929 / 921
NH ₃	115.6	274.4 / 292.3	282.6 / 298.0	0.061	1504 / 1500
HFC-125	117.3	274.1 / 292.5	282.3 / 298.0	0.053	1582 / 1578
HFC-152a	117.8	274.0 / 292.5	282.3 / 298.0	0.053	419 / 418
HFC-41	115.4	273.9 / 292.5	282.3 / 298.0	0.060	1148 / 1141

3.3 Simulation results – simulation results including velocities inside the skylight

3.3.1 Carbon dioxide

Ansys Fluent simulation results obtained with CO₂ inside the skylight are given in Figure 2 for the temperatures and heat fluxes, respectively. This shows the lower temperatures of the LW transmissive CWG window sections and the thermal radiation heat fluxes which are limited to the CWG glass.

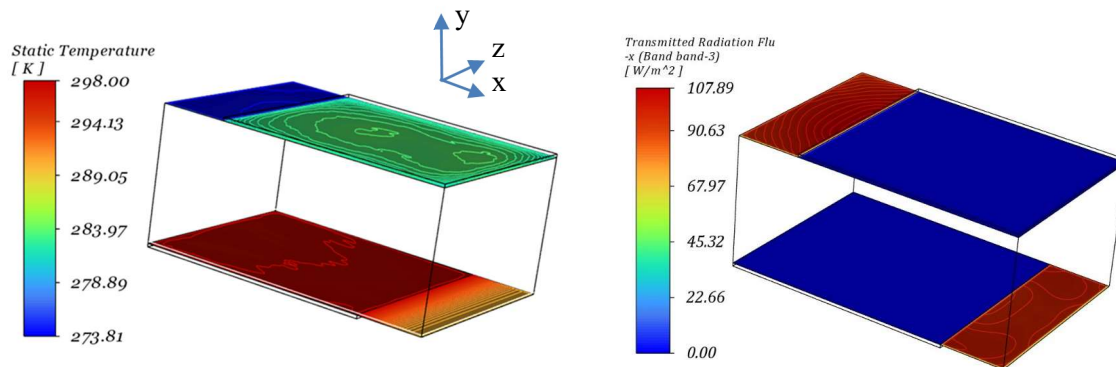


Figure 2: Temperatures (left) and thermal radiation heat fluxes (8-14 μm) (right) for the skylight filled with carbon dioxide, CO₂.

The convection velocities of the gas inside the skylight are shown in Figure 3, as three velocity vector components v_x , v_y , v_z (m/s) as well as the velocity magnitude $|\underline{v}| = (v_x^2 + v_y^2 + v_z^2)^{1/2}$. Here, x is the horizontal direction along the long side, y is the vertical direction upwards, and z is the horizontal direction along the short side of the skylight. All are given for a diagonal surface through the skylight.

Visible from especially the x-direction velocity as well as the vector magnitude is the convective flow “loop” from the lower right side of the skylight to the upper left side; the figure for the y direction shows the upwards flow on the right side inside the skylight and the downwards flow on the left side inside the skylight.

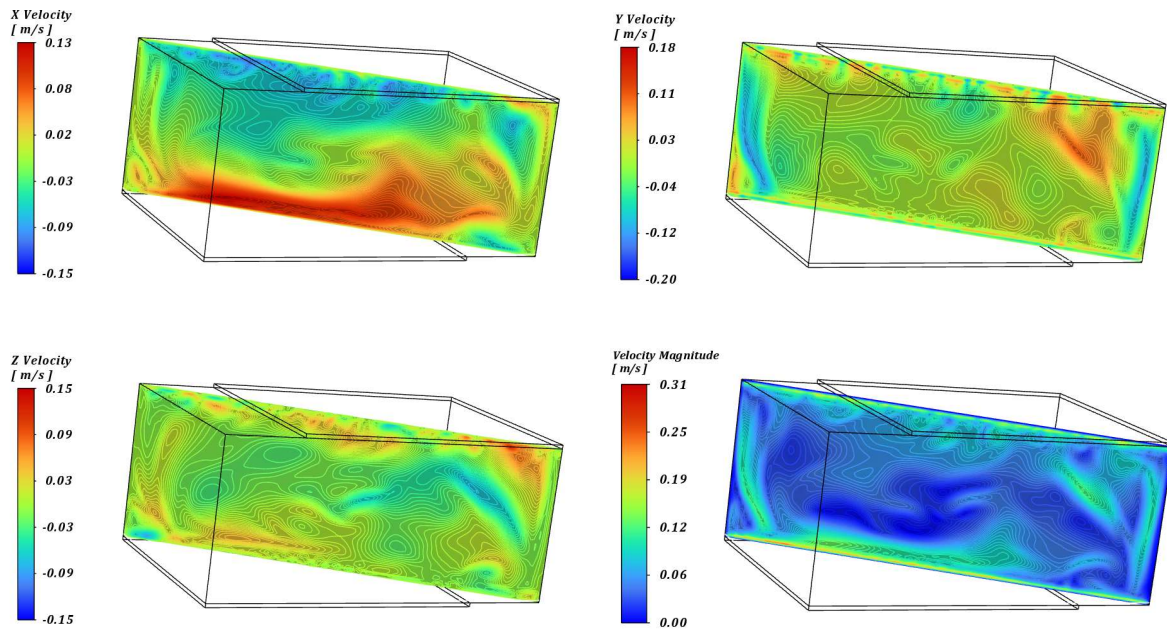


Figure 3: Velocities in x (left, top), y (right, top), z (left, bottom) and as velocity vector magnitude (right, bottom) for the skylight filled with carbon dioxide, CO₂.

3.3.2 Ammonia

The analysis for CO₂ was extended to ammonia, NH₃, a stronger participating gas with absorption bands inside the atmospheric window wavelength range – see Figures 4 and 5. While the volumetric heat content (W/m³), see Table 2 is significantly higher (almost 1½ ×) than CO₂, the convection velocities are similar. The passive cooling effect is around 13% higher (115.6 W/m² vs. 101.9 W/m²) than with CO₂. Strong upwards (y direction) flow is seen above the lower CWG, showing a strong downwards flow below the upper CWG as well.

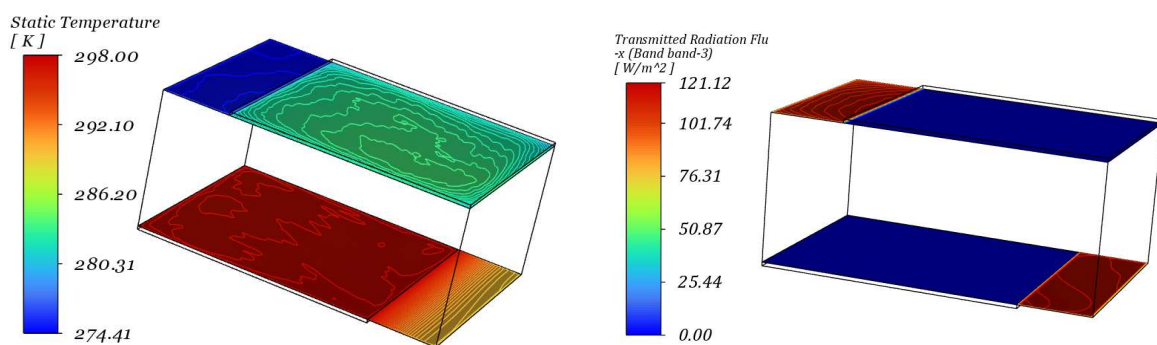


Figure 4: Temperatures (left) and thermal radiation heat fluxes (8-14 μm) (right) for the skylight filled with ammonia, NH₃.

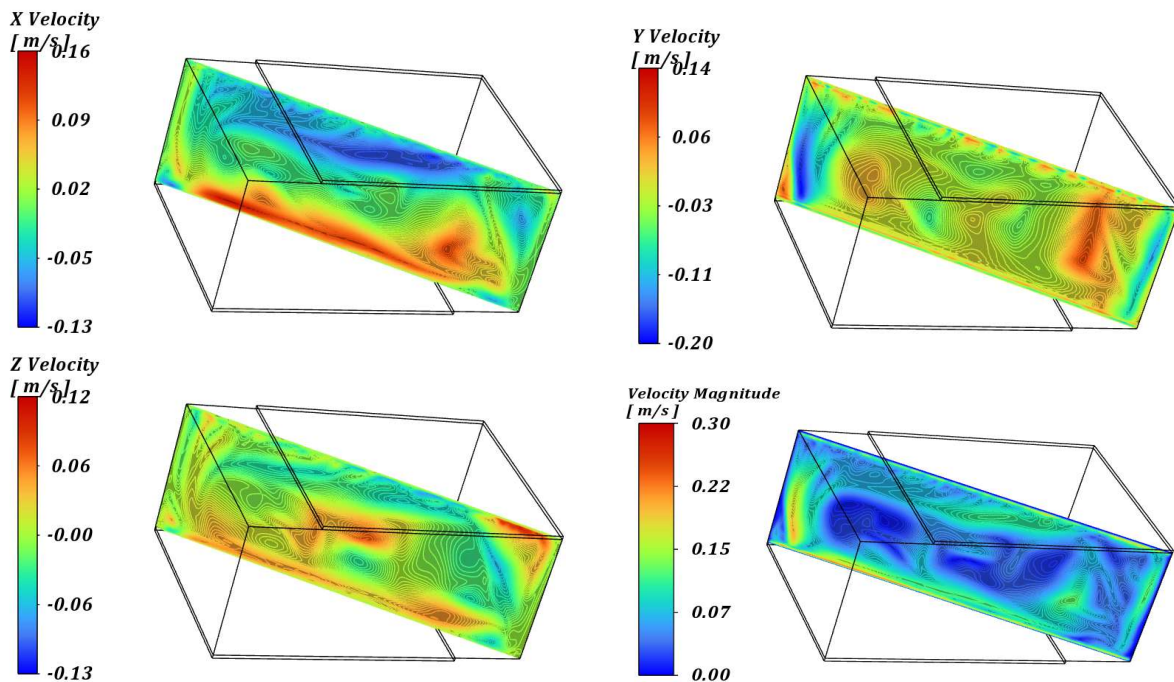


Figure 5: Velocities in x (left, top), y (right, top), z (left, bottom) and as velocity vector magnitude (right, bottom) for the skylight filled with ammonia, NH_3 .

3.3.3 HFC-152a

Since 1,1 - difluoro ethane, HFC-152a may be a suitable alternative for NH_3 , giving a (8-14 μm) passive cooling effect similar to HFC-125 while having a GWP < 150, this is presented here as the third case. Of the four cases given here with graphics – see Figures 6 and 7 – this gas gives the smallest volumetric heat content (419 W/m^3) transported of all six gases considered. Nonetheless, the passive cooling of 117.8 W/m^2 is practically identical to 117.3 W/m^2 obtained with HFC-125. Compared to CO_2 and NH_3 , velocities inside the skylight are smaller, which all things combined, suggests a highly effective transport medium for the application here aimed at. Further studies may show how a smaller volumetric heat content and somewhat lower velocities do nevertheless give the highest passive cooling effect of 18% higher than when using air.

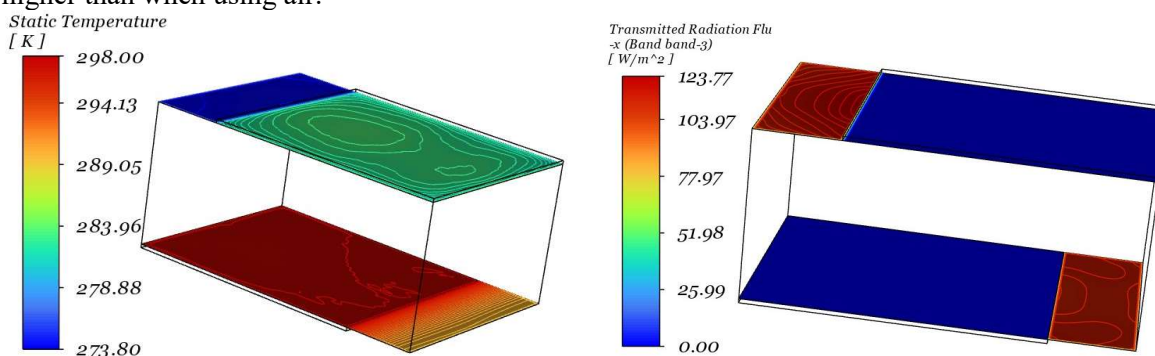


Figure 6: Temperatures (left) and thermal radiation heat fluxes (8-14 μm) (right) for the skylight filled with 1,1 - difluoro ethane, HFC-152a.

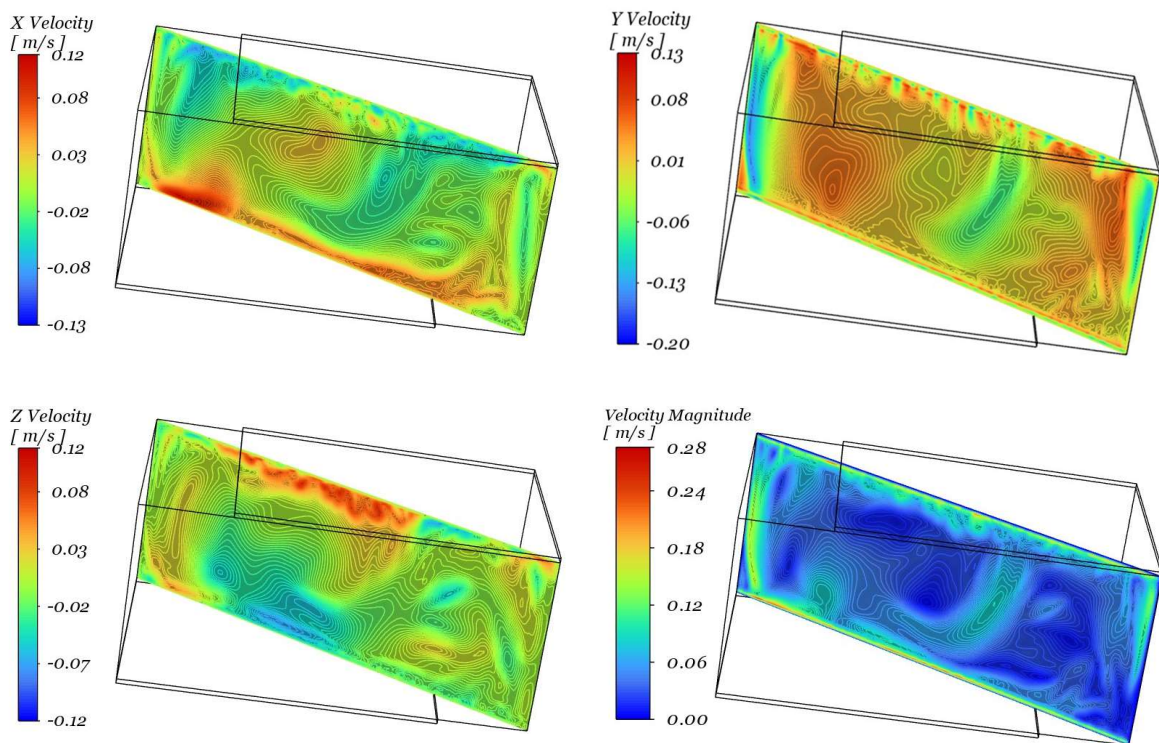


Figure 7: Velocities in x (left, top), y (right, top), z (left, bottom) and as velocity vector magnitude (right, bottom) for the skylight filled with 1,1 - difluoro ethane, HFC-152a.

3.3.4 HFC-41

Finally, also results with fluoro methane, HFC-41 are given here, giving the lowest, 273.9 K, upper CWG temperature, only a fraction lower than the 274.0 – 274.4 found for the other five gases. Volumetric heat amounts absorbed / emitted are larger than for air, CO₂, and HFC-152a, but lower than for NH₃ and HFC-125. Similar to the case with CO₂, the x-direction velocities are relatively high at the lower surface of the skylight.

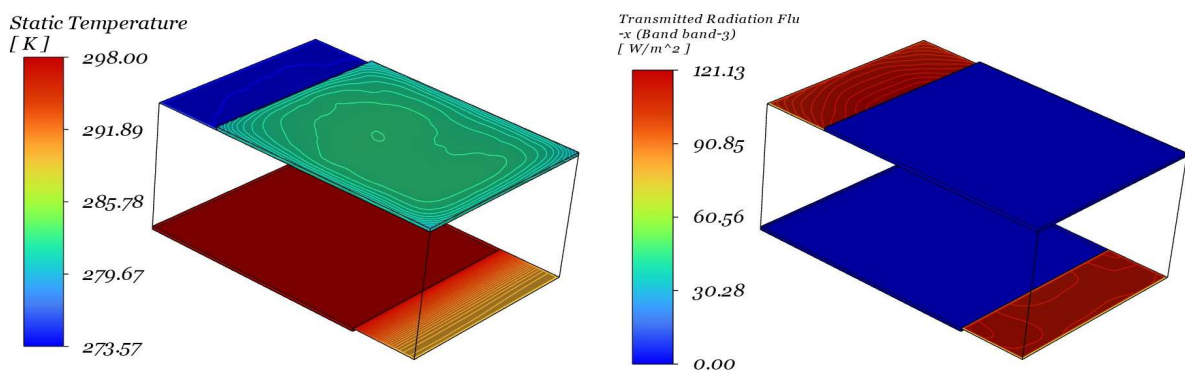


Figure 8: Temperatures (left) and thermal radiation heat fluxes (8-14 μm) (right) for the skylight filled with fluoro methane, HFC-41

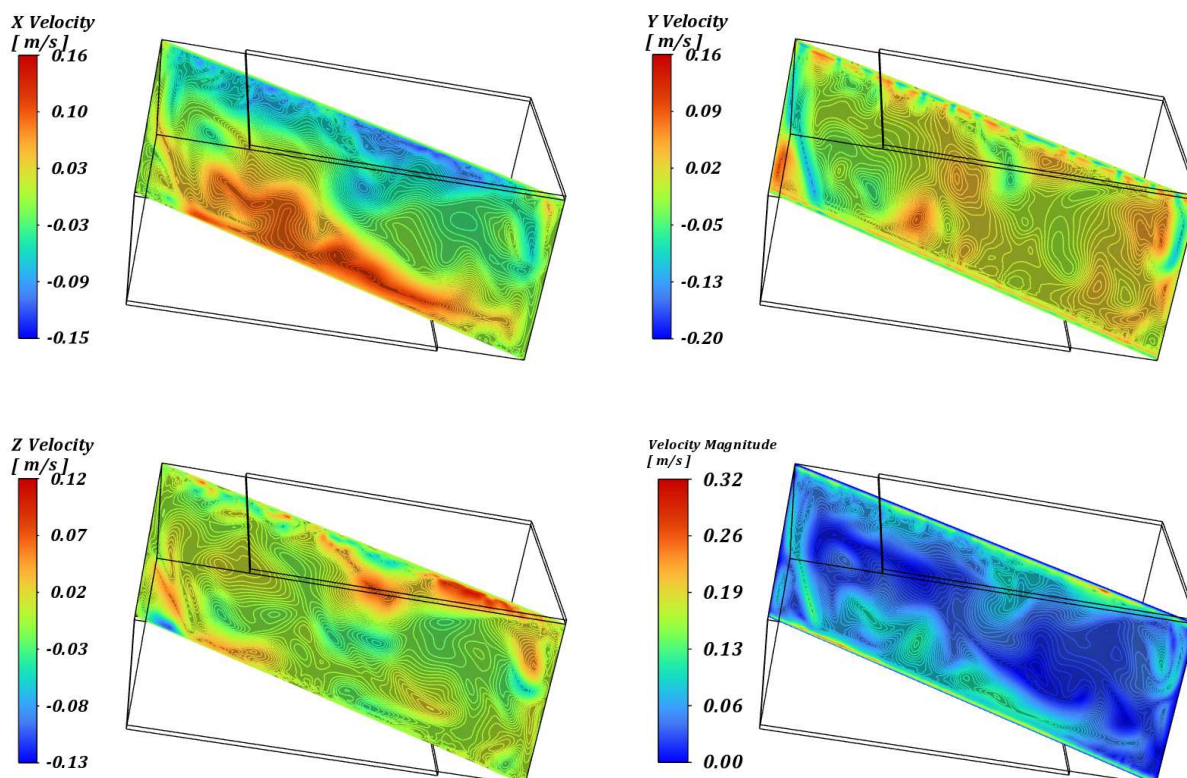


Figure 9: Velocities in x (left, top), y (right, top), z (left, bottom) and as velocity vector magnitude (right, bottom) for the skylight filled with fluoro methane, HFC-41

4 Conclusions

A comparison is given for six gases with thermal radiation participating properties that may be applied inside a passive cooling skylight. While air and CO₂ give a modest effect, an alternative is needed for HFC-125 because of too high GWP for refrigeration systems (incl. passive cooling skylights) and the somewhat unpleasant NH₃. Two low GWP < 150 gases HFC-125a and HFC-41 assessed here using emissivity properties analysis for four thermal radiation wavelength bands, with focus on the 8-14 μm atmospheric window, followed by CFD simulations. The passive cooling flows and the convection flow fields inside the skylight were obtained for a given skylight design. The results suggest that HFC-152a gives better passive cooling (117.8 W/m²) performance than HFC-41 (115.4 W/m²) being similar to the performance with NH₃ (115.6 W/m²) and HFC-125 (117.3). While for HFC-152a this is slightly better than HFC-41, HFC-152a also involves smaller convective flow velocities and the lowest volumetric heat absorption/emission of all six gases assessed and compared. Special for HFC-41 is a high SW radiation emissivity: the consequences of this need further study. At this point HFC-152a seems to be the best alternative for NH₃ and HFC-125. This is part of developments of making the ÅA skylight effective also during daytime – results on various geometries and designs for the skylight are (and have been) reported elsewhere.

Nomenclature

A	Absorbance, -
AC-HP	Air conditioners and heat pumps
c	Concentration of active gas species, mol/mol
CWG	Cleartran® ZnS window glass
GWP	Global warming potential

HVAC	Heating, ventilation and air conditioning
HFC	Hydrofluorocarbon
IR	Infrared
L	Optical path, m
LW	Long wavelength
R-	Refrigerant
SW	Short wavelength
SWG	Standard window glass
v	Velocity, m/s
x,y,z	Cartesian coordinates
α	Absorptivity, -
α'	Molar absorptivity, mol/(mol·m)
ε	Emissivity, -
κ	Absorption coefficient, 1/m
ρ	Reflectance, -
τ	Transmittance, -

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