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Compatibility of elastomers with hydrocarbons – a closer look

Margrit Junk, Joachim Germanus

Short-chain hydrocarbons are widely used as fuels, refrigerants and starting materials for the synthesis of other chemicals. In particular, the use of hydrocarbons as natural refrigerants has been increasing in recent years. Wherever the fluids are stored, transported, processed, or utilized, reliably sealing systems are required. Due to the high flammability of hydrocarbons, stringent requirements must be met regarding tightness and leak prevention. As part of a research project, the performance of elastomer sealing materials in both gaseous and liquid hydrocarbons was investigated through *in-situ* monitoring of their swelling behaviour. The study covered short-chain hydrocarbons ranging from methane to butane.

1 Introduction

Hydrocarbons are increasingly used in refrigeration and air-conditioning systems and heat pumps, providing an alternative to halogenated refrigerants. They often offer higher volumetric cooling capacities than non-flammable fluorinated refrigerants and are generally more environmentally friendly. However, their flammability necessitates stringent safety measures, making reliable sealing of the systems crucial for leak prevention and safe operation.

Today, elastomeric materials are indispensable components of sealing systems, serving in both static and dynamic applications. Their ability to maintain leak-tight operation is essential for ensuring the safety, reliability, and efficiency of refrigeration and air-conditioning systems.

The selection and use of suitable elastomeric materials are crucial, as individual elastomers are only compatible with specific operating conditions and applications. Therefore, the material properties must be carefully matched to the requirements of the respective system and the media in contact with the sealing elements.

Therefore, reliable testing methods are needed to evaluate the properties of materials in these media. The swelling behavior of elastomers is a key parameter for material characterization, as it has a significant impact on sealing performance.

2 In-situ testing of elastomeric materials in hydrocarbons

2.1 *Experimental setup and safety aspects*

The *in-situ* tests were performed in a temperature-controlled autoclave with a sight glass. (Figure 1). To provide the necessary illumination for a visual evaluation of the sample, the autoclave was equipped with a pressure- and fluid-resistant interior light source.

To enable the safe testing of hydrocarbons, a pre-existing test system was modified and adapted to the specific requirements of these media. The electrically driven stirring system of the autoclave was replaced by a pneumatic one to eliminate potential ignition sources. In addition, an extraction enclosure was constructed around the autoclave. Air is continuously extracted from the enclosure and discharged outdoors. Consequently, in the event of a leak, any released hydrocarbon gases are diluted by the incoming air, preventing the formation of a flammable or explosive atmosphere within the enclosure. Several sensors were installed to monitor the atmosphere for flammable gases, both inside the enclosure and in the surrounding laboratory area. Figure 1 shows the test equipment in its operational configuration after completion of the retrofit.

During the experiments, the autoclave temperature was maintained by a thermostatically controlled heating unit. The temperature inside the autoclave was measured with a Pt100 resistance thermometer. The pressure was monitored with both an analogue manometer and a digital pressure sensor. Pressure and temperature were both recorded using a data acquisition system during the experiments.

For the experiments, the O-rings were placed on an adjustable holder that allows a defined degree of elongation of the samples. The holder can be rotated when placed in the autoclave, allowing precise alignment and positioning of the O-rings.

The time-dependent dimension changes of the samples were recorded using a high-resolution camera system. Depending on the fluid density and the changing refractive index, the camera was focused manually. The recorded single pictures were analysed with image processing software. For each sample, the cross-section diameter of the O-ring cords as well as the distance between the cords were measured (Figure 2). To correct for measurement deviations arising from variations in the refractive indices of the test fluids, the sample holder was used as an internal calibration scale for the image-based measurements.

2.2 *Samples and test conditions*

HNBR and NBR were selected for the investigations due to their widespread use as sealing materials and their generally good compatibility with hydrocarbons.



Figure 1: Autoclave system with housing

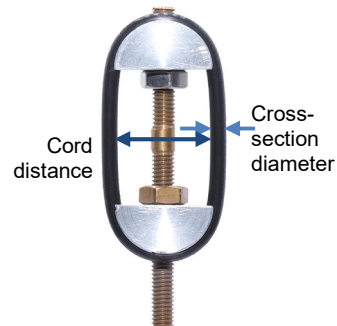


Figure 2: Sample holder

The investigations were performed with 20 x 2 mm O-rings. Before starting the tests, the samples were stored under standard climate conditions. The cross-section diameter and the cord distance of the O-rings were determined in a pre-stressed state under 5% elongation in a sample holder, which was screwed into the autoclave via a pressure-tight connection. The preload was kept constant in all experiments to ensure secure positioning of the samples and to prevent them from slipping off the holder.

The experiments were carried out in both gaseous and in liquid hydrocarbons. The scope of this paper is restricted to the effects of gaseous hydrocarbons. Two basic series of experiments were conducted: one investigating the dependence of volume on hydrocarbon chain length, and another examining the dependence on fluid density.

During the tests, the samples were exposed to the hydrocarbons and observed for periods of typically up to 48 hours. Due to the diffusion- and relaxation-controlled kinetics of swelling, the thermodynamic equilibrium (steady state) is generally reached a few hours after the target temperature and pressure are attained. Accordingly, the test duration was selected to ensure that both the initial rapid swelling phase and the subsequent slower approach to equilibrium could be fully captured. The duration of the relaxation process after pressure release, however, depends on the type of fluid and may extend over several days.

3 Dependence of volume change from hydrocarbon chain length

3.1 Test conditions

To evaluate the dependence of volume change on molecule size, i. e. the chain length of the hydrocarbons, experiments were conducted under isothermal and isopycnic conditions. The parameters were selected such that, at comparable temperatures, the gas density was as similar as possible. As a result, gas pressure varies over a wide range. The details of the experiments are given in Table 1. Most tests were performed at 70 °C, except those with n-butane. At a temperature of 70 °C and a density of 0.025 g/cm³, n-butane exists in both the liquid and gaseous phases. Therefore, the test temperature had to be increased to 80 °C.

For evaluation of the correlation between volume change and hydrocarbon chain length HNBR was used.

Table 1: Test conditions for the dependence of volume from chain length of hydrocarbons

	Methane (R50)	Ethane (R170)	Propane (R290)	n-Butane (R600)	Isobutane (R600a)
Temperature [°C]	70	70	70	80	70
Pressure [bar]	43	20	13.3	10	10
Fluid density [g/cm ³]	0.025	0.023	0.025	0.025	0.025

3.2 Results

With increasing chain length (from methane to butane), the change in cross section diameter of the O-rings increases (see Figure 3 and Figure 4). Additionally, a kinetic dependence can be observed, whereby the equilibrium state of swelling is reached more quickly for smaller hydrocarbon molecules.

The volume change after reaching a stable state after the pressure release is slightly increased with the chain length of the tested hydrocarbons (Figure 4).

The influence of the molecular geometry on swelling behaviour was investigated in comparative tests with n-butane and isobutane. The equilibrium volume change is very similar for both gases. However, n-butane reaches steady state more quickly, as does the recovery of the initial volume after aging (Figure 5). This effect is attributed to steric hindrance, which is assumed to cause isobutane molecules to be retained within the polymer structure of the O-rings.

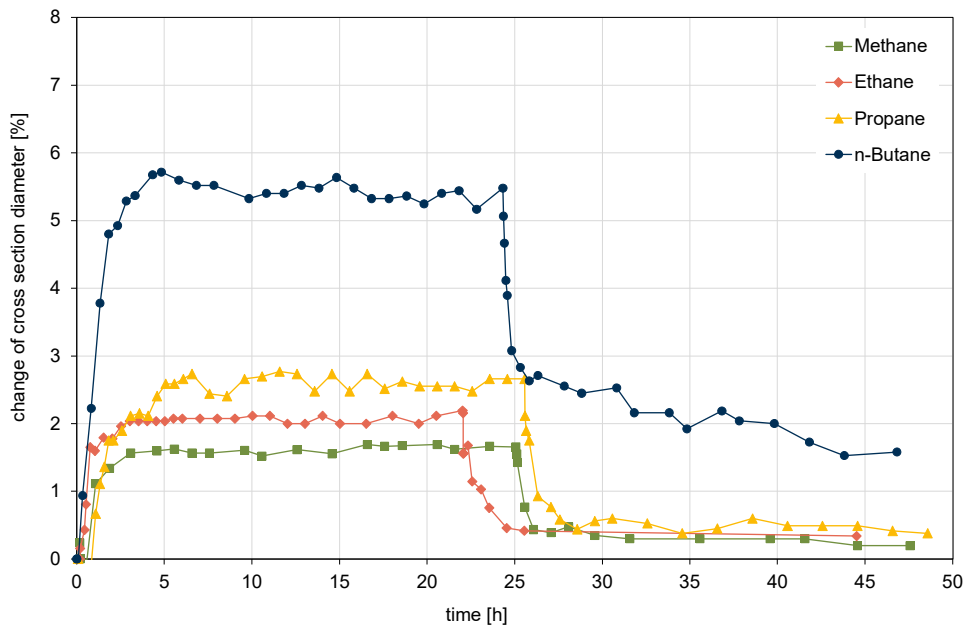


Figure 3: Time dependent volume change of HNBR in different hydrocarbons

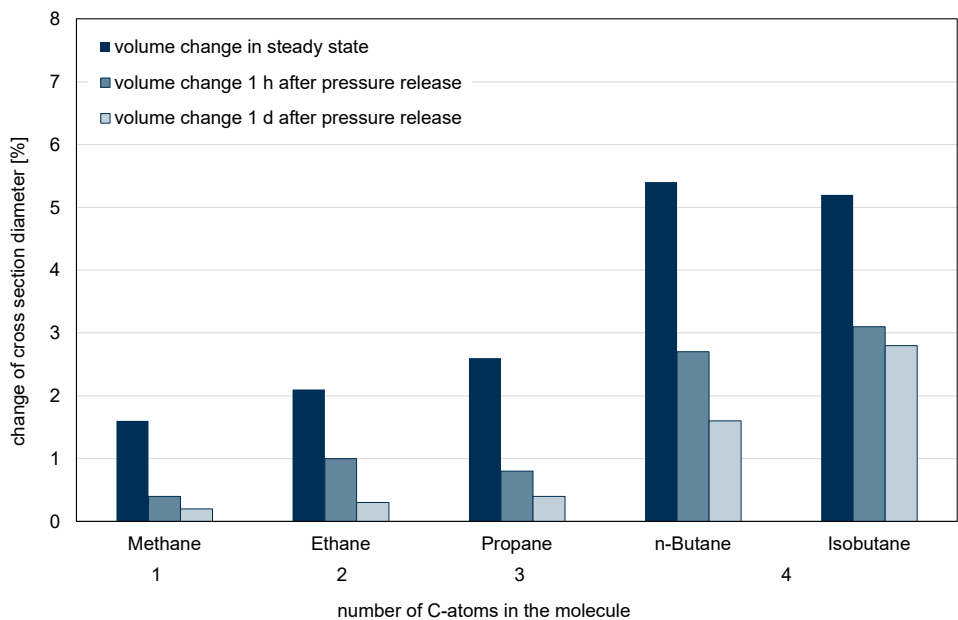


Figure 4: Volume change of HNBR in steady state and after pressure release for different hydrocarbons

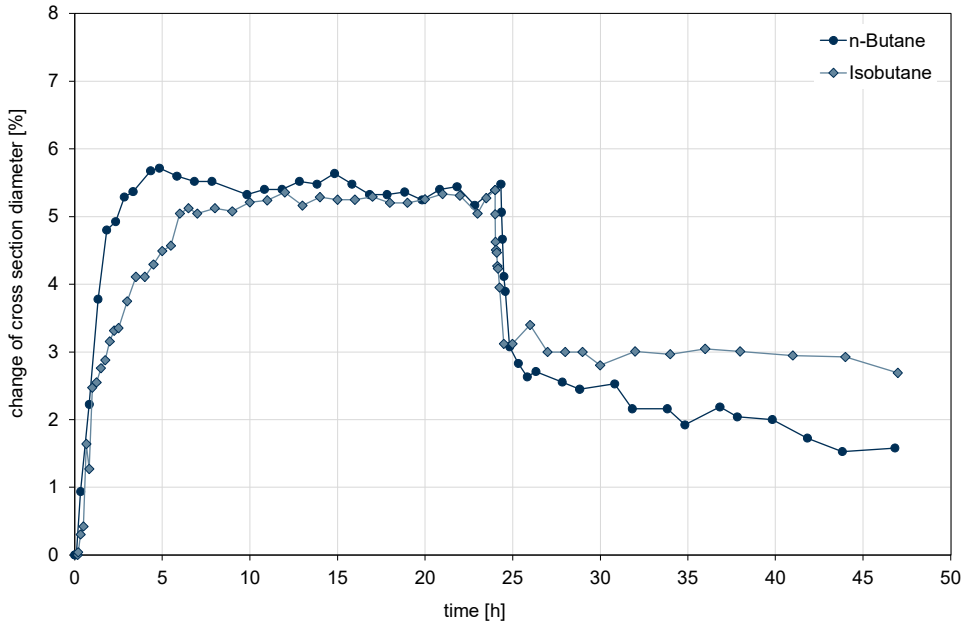


Figure 5: Time dependent volume change of HNBR in n-butane and isobutane

4 Dependence of volume change from gas density

4.1 Test conditions

The test series to evaluate the dependence of volume change on fluid density were conducted with propane (R290) and isobutane (R600a), which are commonly used refrigerants. The test conditions are summarized in Table 2. All experiments were performed under isothermal conditions, with the gas density adjusted via the applied pressure.

The results for experiments with HNBR are presented in this paper.

Table 2: Test conditions for the dependence of volume from density of hydrocarbons

	Propane (R290)			Isobutane (R600a)		
Temperature [°C]	70	70	70	80	80	70
Pressure [bar]	10	13	19	9	10	10
Fluid density [g/cm ³]	0.017	0.025	0.038	0.021	0.024	0.025

4.2 Results

With increasing gas density, the volume change of the HNBR O-rings increases (Figure 6). For larger molecules the effect appears to be more pronounced. Similar effects were observed in a previous project involving the refrigerants R744 (CO₂) and R1234yf.

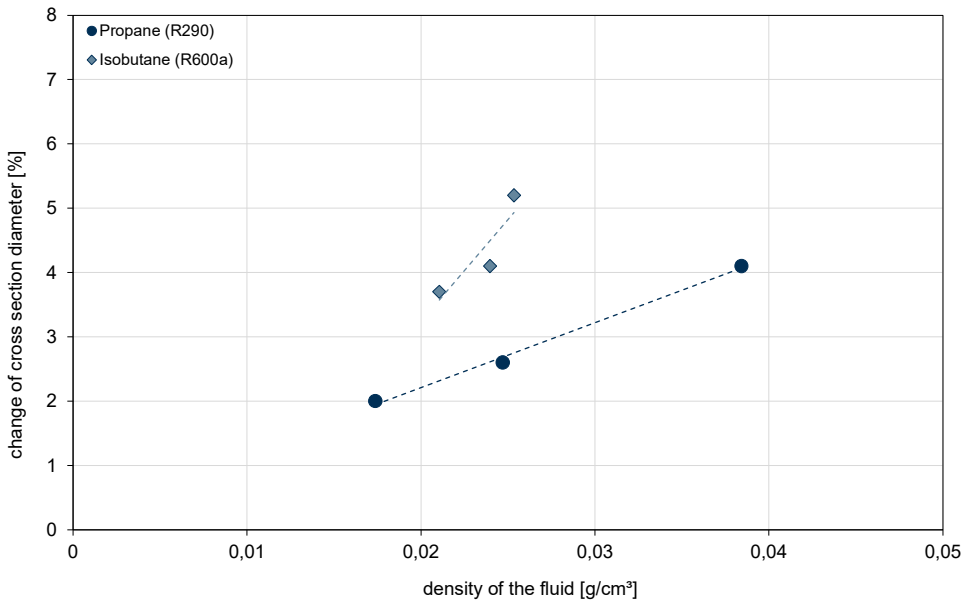


Figure 6: Volume change of HNBR in dependence of fluid density

5 Summary and conclusion

Although the project on which this publication is based focuses on natural refrigerants, the results are also relevant to other hydrocarbon applications and may be transferred accordingly.

The experiments have shown that HNBR and NBR swell upon exposure to hydrocarbons. Already 10 minutes after pressure release, these changes are significantly less pronounced than under fluid contact. This indicates that these effects are not captured in conventional autoclave tests.

Two main factors governing volume change were identified. First, a dependence of volume increase on hydrocarbon chain length was observed. This applies not only to equilibrium swelling but also to permanent changes after removal of the fluid. Second, increasing density of gaseous hydrocarbons leads to increasing elastomer

swelling. Both effects should be considered in the design of seal and the groove geometries.

The data generated in the project contribute to a better understanding of the elastomer behaviour in hydrocarbons and, where sufficient information is available, to improve predictability.

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7 Nomenclature

- (R50) Methane (refrigerant number ac. to ANSI/ASHRAE 34-2022)
- (R170) Ethane (refrigerant number ac. to ANSI/ASHRAE 34-2022)
- (R290) Propane (refrigerant number ac. to ANSI/ASHRAE 34-2022)
- (R600) n-Butane (refrigerant number ac. to ANSI/ASHRAE 34-2022)
- (R600a) Isobutane (refrigerant number ac. to ANSI/ASHRAE 34-2022)

8 Authors

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