Dielectric properties of cryogenic gas mixtures for superconducting power applications

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Abstract. Dielectric breakdown strengths of ternary mixture of gaseous helium, hydrogen, and nitrogen at room temperature and cryogenic temperatures (77 K) have been investigated experimentally in an effort to identify a gaseous cryogen with higher dielectric strength than pure helium for high temperature superconducting (HTS) power applications. Building on our previous success in enhancing the dielectric strength of helium gas by 80\% by using a binary mixture containing 4 mol\% hydrogen + 96 mol\% helium, a ternary mixture with a composition of 4 mol\% hydrogen + 88 mol\% helium + 8 mol\% nitrogen was investigated. The composition of the ternary mixture was the result of theoretical modelling studies which used the Boltzmann equation to calculate the dielectric strength of various ternary mixture ratios. The dielectric breakdown strength of the ternary mixture was measured at room temperature and at 77 K at several pressures up to 2 MPa. A 300 \% enhancement in dielectric strength was observed for the ternary mixture compared to pure gaseous helium. The dielectric strength of the ternary mixture was measured to be 12 kV/mm at 1 MPa and 77 K, which is approaching the dielectric strength range of liquid nitrogen. The results on the new mixture are compared to the previous studies on helium-hydrogen binary mixtures and the implications of the observed enhancement for gas cooled HTS power devices is discussed. The challenges posed by the addition of nitrogen in terms of limited operating temperature and pressure due to the condensation of nitrogen at higher pressures and/or lower temperatures is also discussed.

1. Introduction
Prototypes of gaseous helium cooled High Temperature Superconducting (HTS) power cables and other high power density devices have been demonstrated [1]. Additional superconducting power applications cooled with closed loop gaseous helium (GHe) circulation have been considered to exploit the higher current carrying capacity of HTS materials at low temperatures (< 77 K) or to utilize superconducting materials such as MgB\textsubscript{2} with lower Tc values [2]–[4]. The US Navy also has a preference in utilizing GHe instead of LN\textsubscript{2} as there is a reduced risk of asphyxiation [5], [6]. One of the challenges posed by GHe is its low dielectric strength which has limited GHe cooled technology to low-medium operating voltages. We have been investigating methods to increase the dielectric strength of GHe to enable higher voltage ratings of GHe cooled superconducting power technology. We demonstrated an 80\%
enhancement of dielectric strength of GHe with the addition of 4 mol% hydrogen [7], [8]. We have continued this research by systematically exploring the options for further enhancing the dielectric strength of GHe. Our ongoing research on this topic includes optimizing the helium-hydrogen mixture to have the greatest improvements in the dielectric properties without safety concerns regarding the flammability limit of hydrogen.

We have also been investigating the dielectric strength of GHe by developing ternary gas mixtures containing hydrogen, helium and nitrogen. The addition of nitrogen gas to the helium-hydrogen mixture was expected to have superior dielectric properties compared to the 4 mol% hydrogen mixture balanced with GHe. Theoretical modelling of ternary mixture containing varying mol% of nitrogen with a fixed mol% of H$_2$ (7 mol%) and balanced with GHe was performed using the Boltzmann equation. The model predicted that the ternary mixture would have a significantly higher dielectric strength than pure helium [9]. Additionally, as both hydrogen and nitrogen have superior heat capacity than helium, the ternary mixture it is expected to have cooling capabilities equal to or greater than GHe. Both the heat capacity and dielectric strength of cryogens need to be taken into consideration when rating the performance of HTS power devices. This paper focuses on the dielectric aspect of the ternary mixture with future work to be focused on the heat capacity.

The theoretical modelling also took into consideration how the addition of N$_2$ gas to the ternary mixture can lead to a reduction of the operating pressure and temperature range capable by GHe due to the possibility of condensation of N$_2$ [9]. Optimization of the N$_2$ mol% was performed to ensure the desirable operating temperatures and pressures could be achieved. A ternary mixture containing 4 mol% H$_2$, 8 mol% N$_2$ and 88 mol% GHe was selected for experimental characterization as the modelling suggested this mixture could operate at 1.0 MPa at 77 K without condensation of N$_2$ occurring. This paper discusses the experimental investigation on AC and DC breakdown of the ternary mixture containing 4 mol% H$_2$, 8 mol% N$_2$, and 88 mol% GHe at both room temperature and at 77 K. The results obtained for the ternary mixture are compared to the AC and DC breakdown measurements previously obtained for the binary mixture consisting of 4 mol% H$_2$ and 96% GHe at room temperature and 77 K. To allow for an accurate comparison between the binary and ternary mixtures breakdown measurements performed on GHe were used as a benchmark for reproducibility and validation.

2. Experimental Methodology

The experimental setup to measure the dielectric properties of the ternary mixture is similar to what has been described in [7], [8] and is a modification of the experimental setup provided by IEEE Standards 4 to measure voltage by means of a gap distance between electrodes. The gap distances and electrode dimension/profile were selected to ensure a uniform electric field and the ability to perform several hundred breakdown measurements without damaging/changing the profile of the electrode. This ensures consistency in the measurements and allows for a useful comparison to be made between various gas mixtures studied. In the experiments the breakdown voltage is determined by the voltage required to arc across a pair of 25 mm diameter stainless steel electrodes which have a similar profile to Bruce and produce an uniform electric field between them [10]. The gap distance between the electrodes was set to 1 mm using a gauge block set and the gap distance was kept constant throughout the experiment. Previously a gap distance of 2 mm has been used in [7], [8], however there was a concern that the 2 mm gap distance in the experiments for the ternary mixture the breakdown voltage would exceed the maximum voltage rating of the high voltage bushing used in the circuit. While the high voltage transformer is rated for 100 kV AC, a VARIAC has been connected in series to the primary side which allows for a greater resolution of the voltage step but reduces the maximum voltage rating of the circuit. This configuration allows for a fixed ramp rate of approximately 100-200 V/s to be achieved which ensures a greater level of accuracy for the results obtained at voltage levels <3 kV.

The electrodes are installed within a pressure vessel, with the top electrode connected to the high voltage source and the bottom electrode grounded (Figure 1). Once the electrode setup is installed within the pressure vessel several vacuuming and flushing cycles are performed to ensure a high purity gaseous environment is obtained. A dry-scroll pump was used to ensure a sufficient vacuum of 2.2×10$^{-2}$ mbar.
was achieved. The flushing cycles were performed using nitrogen and industrial grade GHe (99.8% purity) at 2.0 MPa at room temperature, with a vacuum cycle between each flushing cycle. The pressure vessel was then filled with 2.0 MPa of research grade GHe (99.9999% purity) at room temperature. Ten AC breakdown measurements were performed to season the electrodes. A set of ten breakdown measurements was sufficient to season the electrodes as after the first 3 measurements the voltage levels recorded were consistent which demonstrated there were no issues with the surface modification of the electrodes. After seasoning the electrodes, a set of 15 AC breakdown measurements were performed. To ensure that the local temperature at the electrodes is 77 K, several minutes of waiting time between successive measurements was built into the measurement protocol. The circuit was then switched to DC configuration by connecting diodes and a capacitor to the AC transformer to create a half wave rectified signal. Again, a set of 15 DC breakdown measurements were performed. The pressure within the vessel was released to 1.5 MPa, then to 1.0 MPa, and finally 0.5 MPa with a set of 15 AC and DC breakdown measurements performed at each pressure level. After the last set of measurement had been performed in pure helium gas medium, the pressure vessel was vacuumed before being filled with 2.0 MPa at room temperature of the ternary mixture. It should be noted that the ternary mixture was provided premixed by a commercial gas supplier to ensure that there was no issue with gas separation during the experiments. The experimental procedure used for pure GHe was repeated to obtain breakdown voltage data for AC and DC at the same pressure levels of 2.0 MPa, 1.5 MPa, 1.0 MPa, and 0.5 MPa for the ternary mixture. After completing the breakdown measurements at room temperature the pressure vessel was fully immersed in LN2 (Figure 2). The pressure within the vessel was increased to 1.0 MPa and a sufficient wait time was included in the measurement protocol to ensure that thermal equilibrium had reached before the measurements at 77 K were performed. The same measurement procedure as outlined above was performed with 15 AC and DC breakdown measurements at 1.0 MPa, 0.85 MPa, 0.75 MPa, 0.5 MPa, and 0.25 MPa. On completion of the last breakdown measurement the dry scroll pump was

Figure 1. The electrode setup with uniform field in the gap between the electrodes [7].
connected to achieve a vacuum of $2.2 \times 10^{-2}$ mbar. The pressure vessel was then flushed with 2.0 MPa of industrial grade GHe at 77 K, the dry scroll pump was reconnected to achieve a vacuum of $2.2 \times 10^{-2}$ mbar, and then the pressure vessel with 1.0 MPa research grade GHe at 77 K. The pumping and flushing cycles in between measuring the ternary mixture and pure GHe ensured the removal of any nitrogen and hydrogen contaminants. The same measurement procedure as outlined above was performed on pure GHe with a set of 15 AC and DC breakdown measurements performed at 1.0 MPa, 0.85 MPa, 0.75 MPa, 0.5 MPa, and 0.25 MPa. On completion of the last measurements at 0.25 MPa, the pressure was increased to 2.0 MPa. Sets of 15 AC breakdown measurements were performed at 2.0 MPa and 1.5 MPa to give additional GHe data points to allow for a comparison between the ternary and binary mixtures to be made.

3. Results

The data from the AC and DC breakdown measurements on pure GHe, the binary mixture (4 mol% H$_2$ and 96 mol% He) and the ternary mixture (4 mol% H$_2$, 8 mol% N$_2$, and 88 mol% He) at pressures between 0.5 MPa and 2.0 MPa at room temperature are shown in Figure 3 and Figure 4, respectively. The error bars in Figure 3 and Figure 4 indicate the maximum and minimum voltage recorded at each pressure level. The data for the binary mixture are included for the purpose of comparison and have previously been presented in [11]. The measurement protocol used for the binary mixture is identical to that used for the pure He and the ternary mixture in this study, and hence the comparison is useful. The data on GHe obtained during the study of the binary mixture were consistent with the measurements on GHe obtained during the study of the ternary mixture, making the comparison of the data on pure GHe, the binary mixture and the ternary mixtures meaningful and useful for drawing conclusions on the effect of additions of H$_2$ and N$_2$ for He gas on the AC and DC breakdown characteristics.
Figure 3. AC breakdown voltage as a function of pressure at room temperature in homogeneous field of pure GHe, binary mixture and ternary mixture.

Figure 4. DC breakdown voltage as a function of pressure at room temperature in homogeneous field of pure GHe, binary mixture and ternary mixture.

The data in Figures 3 and 4 demonstrate the benefit of introducing N₂ as the third component in the ternary mixture in enhancing the dielectric strength. The dielectric strength of the ternary mixture represents a 300% enhancement in the dielectric strength of GHe. This is significantly greater than the 80% enhancement dielectric strength obtained by the addition of 4 mol% H₂ as in the binary mixture.
Figure 5. AC breakdown voltage as a function of pressure 77 K in homogeneous field of pure GHe, binary mixture and ternary mixture.

Figure 6. DC breakdown voltage as a function of pressure at 77 K in homogeneous field of pure GHe, binary mixture and ternary mixture.

The data of breakdown measurements of the GHe, the binary mixture, and the ternary mixture between 0.25 MPa and 1.0 MPa at 77 K for AC and DC voltages are shown in Figure 5 and Figure 6 respectively. The error bars in Figure 5 and Figure 6 indicate the maximum and minimum voltage recorded at each pressure level. For the ternary mixture, measurements were only performed up to 1.0 MPa because of the potential for condensation of nitrogen at higher pressures, whereas the
measurements for GHe were performed up to 2.0 MPa to allow a comparison to be made to the previous breakdown data collected for GHe using a 2 mm gap distance [8]. Similar to the room temperature data, the values obtained for GHe during the two separate studies of the binary mixtures and ternary mixture are comparable, allowing for a useful comparison to be made between the two data sets.

Similar to the data from the room temperature measurements, the measurements performed at 77 K demonstrate a significant enhancement of dielectric strength in the range of 350–400% of the ternary mixture compared to that of pure GHe. Figures 5 and 6 suggest that the dielectric strength of the ternary mixture plateaus as it reaches 1.0 MPa. The large variation in measurements at 0.75 MPa, 0.85 MPa, and 1.0 MPa for both AC and DC cases suggests condensation of some nitrogen gas at these pressure levels. The dielectric strength values obtained for the ternary mixture at 0.25 MPa and 0.5 MPa do not appear to have been affected by the condensation of N₂ gas. The breakdown voltages obtained for these pressure levels are similar to the what was obtained by the ternary mixture at room temperature at 1.0 MPa and 2.0 MPa, when normalized by the density increase from room temperature to 77 K. The gas density at 1.0 MPa and 2.0 MPa at room temperature is approximately equivalent to that at 0.25 MPa and 0.5 MPa at 77 K, and therefore similar breakdown voltage values are expected.

4. Discussion
The results of the experiments on the ternary mixture discussed in the earlier section and the comparison of the dielectric strengths of pure helium gas, the binary mixture and the ternary mixture demonstrate the utility of gas mixtures in significantly enhancing the dielectric strength of pure GHe for superconducting electric power applications. The dielectric strength of the ternary mixture containing 4 mol% H₂, 8 mol% N₂ and 88 mol% He is approximately 11 kV/mm at 0.85 MPa and 77 K, high enough to be comparable with the dielectric properties of liquid nitrogen (between 12 and 50 kV/mm at pressure levels between 200 kPa and 500 kPa) at 77 K [12]–[15]. It should be noted that further research is still required in determining the exact temperature and pressure ranges of operation that would eliminate the possibility of partial condensation N₂ gas in the mixture. For future experiments it may be beneficial to perform the measurements in ascending pressure levels rather than descending pressure levels. This would allow for a better understanding of when the N₂ within the ternary gas mixture will begin to condense and how the condensation affects the dielectric strength of the gas.

Caution also needs to be taken when developing mixtures with N₂ to ensure that the addition of the gas does not affect its practical application. While enhancing the dielectric strength of GHe is important it is only one aspect of a superconducting power device. One of the major benefits of using GHe in place of LN2 as the cryogen is the larger temperature operating range which allows for higher current densities to be achieved. The addition of N₂ to GHe effectively limits the lowest operation temperature of the device. The other restriction placed on gas mixtures containing N₂ is the maximum operating pressure. The heat capacity of GHe is relatively low and high operating pressures are required to achieve the needed volumetric heat capacity and the required mass flow rate, which is a linear function of the gas density, and hence the pressure. For the 30 m long HTS cable demonstration at the Center for Advanced Power Systems, mass flow rate in the range of 8-10 g/s was obtained by operating the cable at 2.0 MPa. The 8-10 g/s mass flow rate was necessary to maintain the temperature gradient across the inlet and outlet of the HTS cable system at < 4 K [16]. It is also important to ensure that the condensation does not block the heat exchangers attached to the cryocooler which tend to be at the lowest temperature in the cryogenic cooling loop. Addition of N₂, is extremely beneficial in addressing the dielectric strength, one of the major challenges of GHe cooled HTS devices. A theoretical study on the dielectric behaviour of gas mixtures over a wide temperature range and a methodology to identify potential gas mixtures and their useful temperature and pressure ranges is currently in progress.

5. Conclusion
A significant enhancement of dielectric strength of gaseous helium was achieved by the addition of hydrogen and nitrogen. The ternary mixture 4 mol% H₂ + 88 mol% He + 8 mol% N₂ exhibited a superior dielectric strength representing a 300% enhancement over pure gaseous helium. The dielectric strength
measurements conducted at room temperature and 77 K confirmed the previously observed linear relation between the dielectric strength and gas density within the pressure range of interest. A comparison of the data at room temperature and 77 K indicated partial condensation of nitrogen at 1 MPa and 77 K. The data suggests that care must be taken when using the gas mixtures and limit the operating temperature and pressure ranges to regions that do not lead to condensation of nitrogen.

6. References

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