

MASTER'S THESIS

DIVISION OF MATERIALS ENGINEERING

March 2019

# Optimizing the sealing of tubular heating elements

**Author:** Victor Almlad

Company: Backer AB

Supervisor: Dmytro Orlov

Co-supervisor: Hans-Göran Künkel

Examiner: Srinivasan Iyengar



**LUND**  
UNIVERSITY

Cover: Image of a tubular heating element.

Master Thesis

ISRN LUTFD2/TFMT--19/5059—SE

Division of materials engineering

Department of Mechanical Engineering

Lund University

Box 118

SE-221 00 Lund

Sweden


© Victor Almblad. All rights reserved.

Printed in Sweden by Media-Tryck, Lund University

Lund 2019



Media-Tryck is an environmentally certified and ISO 14001:2015 certified provider of printed material. Read more about our environmental work at [www.mediatryck.lu.se](http://www.mediatryck.lu.se)

**MADE IN SWEDEN** 

# Populärvetenskaplig sammanfattning

## Optimering av rörelement

*Victor Almblad*

**Rörelement tillverkade av Backer AB används idag som uppvärmningssystem inom bland annat fordons-, medicinteknisk- och hushållsindustrin. Vid användning går en elektrisk ström genom elementet vilket avger värme till omgivningen. För att undvika läckström används magnesiumoxid inuti rörelementen var låga elektriska ledningsförmåga gör den till en utmärkt isolator. Magnesiumoxiden är hygroskopisk och kan enkelt absorbera vatten från omgivande luft. Detta är ett stort problem då det leder till sämre elektrisk isolation inuti elementen. För att undvika detta så försluts rörelementen vilket skyddar magnesiumoxiden och förhindrar inträngande av fukt.**

I produktion är magnesiumoxiden initialt ett hygroskopiskt pulver som efter valsning (pressning) övergår till ett fast, keramliknande hölje inuti elementen. Detta hölje omger en resistansstråd vilken leder ström och avger värme när elementet är aktivt. I kontakt med vatten (fukt), övergår den hygroskopiska magnesiumoxiden till magnesiumhydroxid, också hygroskopisk. Magnesiumhydroxid har förmågan att binda vatten vilket leder till en ökad elektrisk ledningsförmåga hos ämnet. När en ström sedan färdas genom motståndstråden ökar således risken för läckström vilket kan leda till överslag i elementet och i vissa fall kortslutning.

För att förhindra detta försluter Backer AB sina rörelement. Förslutningsmaterialet varierar och beror på vilken miljö som rörelementet skall utnyttjas i. Egenskaper som vidhäftningsförmåga, ogenomtränglighet av vatten och robusthet kan därför bli olika viktiga och behöver tas i beaktning när förslutningsmaterialet väljs. De flesta förslutningar behöver härda och härdningstid har blivit en viktig faktor som påverkar produktionstiden av rörelement.

I detta projekt har den elektriska resistansen hos rörelement undersökts då de utsätts för miljöer med olika luftfuktighet och temperatur. Resultaten visar att utan förslutning så kommer fukt att tränga in i rörelementen vilket leder till en betydligt sämre elektrisk isolation av elementen. Att undvika fukt i produktionen är en stor utmaning och Backer AB strävar mot ökad en förståelse kring vilka faktorer som orsakar dålig elektrisk isolation i rörelementen.

Det finns även ett strakt intresse att förbättra förslutningsprocessen men även att finna nya lämpliga förslutningsmaterial. Många förslutningsmaterial idag kräver lång härdningstid samt värme. Vissa förslutningsmaterial kan även orsaka allergi vilket ställer tuffa krav på hantering och utrustning. Att undersöka och finna nya metoder och/eller ämnen som kan användas vid förslutning är idag ett stort intresse för Backer AB och i detta projekt har bland annat UV-härdning undersökts som ett alternativ för att erhålla en kortare härdningstid. Utöver detta har litteratur och studier undersökts för att finna material som kan komma att användas för att förbättra förslutningar av rörelement.

För intresserade finns mer information att tillgå i ***Optimizing the Sealing of Tubular Heating Elements***.

# Popular science summary

## Optimization of tubular heating elements

*Victor Almlad*

**Tubular heating elements are widely used in automotive, medical and household industries. Today, the use of magnesium oxide (MgO) as electrical insulating material prevents current leakage in heating elements. The flip side of such use is the sensitivity of heating elements to moisture. This is a major problem necessitating the insulation of heating elements by adding sealing components.**

When an electric current is applied to a heating element, a heating wire in its core heats up, and generated heat is transferred to a mantle at the element periphery through an insulation material. The insulation material should be blocking the conduction of electric current from the heating wire to the mantle while conducting heat with minimum losses. This enables efficient heating of surrounding medium, which is the main purpose of a heating element operation. MgO is used as the insulating material in the form of a solid powder. In the presence of moisture, MgO reacts and converts into magnesium hydroxide, Mg(OH)<sub>2</sub>. If hydrated, the Mg(OH)<sub>2</sub> becomes electrical conductive, which generates the risk of current leakage or even short circuit in a worse scenario.

To prevent the penetration of moisture into heating elements a sealing is necessary. The sealing material can vary, and often depends on the conditions where the element is used. Various properties such as adhesion, sealing ability and durability need to be considered when choosing the sealing material. Most sealing materials also require curing before such properties are achieved. Curing is an important step in the sealing process, and the time of curing can be influenced by the source of curing and the choice of material itself.

In this project, investigation of the dependence of electrical insulation where heating elements are exposed to humid conditions has been performed. MgO being hygroscopic both attracts and absorbs moisture from the ambient air. Once moisture is absorbed, MgO converts into Mg(OH)<sub>2</sub>, which causes bad electrical insulation of the heating element, especially when hydrated. The results of this project demonstrate the importance of sealing. Further study revealed the importance of environmental control in production, as exposure to ambient environment has major influence on decrease in electrical resistance. This becomes very important after heat treatment since elevated temperature stimulates the conversion of MgO to Mg(OH)<sub>2</sub>.

This proves the necessity of sealing in order to protect MgO and to prevent the formation and hydration of Mg(OH)<sub>2</sub>. Many sealing materials used today require both significant time and energy and there is a major interest in the improvement of sealing and sealing process. New UV-curable resin materials and various ways of curing were tested in this project. UV-curing demonstrated great promise in reducing the time of curing and new resins were capable of preventing the moisture penetration to samples under very humid conditions. Still, further improvement and investigation is needed regarding the temperature range and efficient curing.

For interested, more information about the subject and challenges is available in the thesis

***Optimizing the Sealing of Tubular Heating Elements.***

## Abstrakt

Rörelement används idag till elektrisk uppvärmning inom flertalet olika industrier, bland annat fordons-, medicinteknik- och hushållsindustrin. NIBE Element är idag det världsledande företaget på produkter och system för elektrisk uppvärmning med huvudkontor Backer AB i Sösdala. Rörelement producerade av Backer AB består huvudsakligen av en motståndstråd, mantel, magnesiumoxidpulver och en förslutning. När en elektrisk ström går genom motståndstråden avges värme till omgivningen utan risk för läckström. Förhindrandet av läckström sker genom utnyttjande av magnesiumoxid (MgO) vars låga elektriska ledningsförmåga gör den till en utmärkt elektrisk isolator.

MgO är hygroskopisk och kan absorbera vatten från dess omgivning (även luft). Vid absorption av vatten bildar MgO magnesiumhydroxid ( $Mg(OH)_2$ ), vilken också är hygroskopisk. Under fuktiga förhållanden absorberar magnesiumhydroxid vatten vilket leder till hög elektrisk ledningsförmåga. Således är fukt ett stort problem vilket gör det nödvändigt att skydda rörelementen med en förslutning. Dagens produktion förhindrar möjlighet till tidig förslutning av rörelementen vilket ökar risken för penetration av fukt i rörelementen.

I detta projekt har den elektriska isolationsförmågan hos rörelement undersökts då rörelement exponeras för olika grader av luftfuktighet och temperatur. Det har även gjorts studier för att undersöka möjligheter gällande nya förslutningsmaterial i framtida produktion. Olika tester har innefattat resistansmätningar av rörelement som exponerats för olika temperatur och luftfuktighet. Penetrationsdjup av fukt inuti rörelement samt möjlighet till kortare härdningstids med hjälp av UV-härdande förslutningsmaterial har också undersökts i detta projekt.

Resultaten från resistansmätningar visade att rörelement är mycket känsliga för fukt, speciellt efter värmebehandling. Detta berodde på att höga temperaturer i produktionen leder till bildande av undertryck vilket gjorde att omgivande luft sögs in i rörelementen. Luftfuktigheten efter värmebehandling visade sig således ha stor inverkan på rörelementens elektriska isolationsförmåga, vilket undersöktes med en klimatkammare.

Resultaten visade även att fukt kunde tränga djupt in i rörelementen men existerade mer koncentrerad vid ändarna. Slutligen var det även möjligt att försluta ett rörelement med kort härdningstid vid utnyttjande av UV härdande förslutningsmaterial. Utmaningar gällande temperaturtålighet och effektiv härdning kräver dock fortsatt arbete innan dessa kan användas industriellt.

**Nyckelord:** Förslutning, värmeelement, luftfuktighet, magnesiumoxid, hydrering, magnesiumhydroxid, isolation, klimatkammare, torr lager, konduktivitet, resistans, vidhäftningsförmåga, härdning.

## Abstract

Tubular heating elements, produced by Backer AB are today commonly used, serving as systems for intelligent heating in automotive, medical and household industries. Tubular heating element consists of a mantle, resistance wire, electrical insulation (magnesium oxide) and a sealing. When active, an electric current transfers heat from the resistance wire of the heating element to the surroundings without the risk of current leakage. This is due to electrical insulation obtained by magnesium oxide (MgO), which possesses low electric conductivity making it an excellent electric insulator.

MgO is hygroscopic and in the presence of moisture, MgO absorbs water and forms magnesium hydroxide ( $\text{Mg(OH)}_2$ ), which if hydrated causes bad electrical insulation of heating elements. This makes moisture a major problem and necessitates insulation through the usage of a sealing. Today, the application of sealing takes place in the latter stages of production, increasing the risk of moisture penetration of the heating elements during fabrication.

In this project, the evolution of MgO electrical conductivity in the presence of moist atmosphere and the effect of insulation of heating elements on this process have been investigated. The heating elements were produced according to company standard, provided for testing by Backer AB. A climate chamber was used to create controlled humid environments, and a dry stock was used for storage of heating elements under investigation. An insulation tester was used to measure resistance, and a UV kit was procured in order to test UV curable adhesives as potential sealing materials. The testing involved the measurement of resistance of samples after storage in humid environment. Depth of moisture penetration, the time effect of sealing process and the sealing of a heating element with UV-curable adhesives were also investigated in the project.

The analysis of experimental data revealed that heating elements are very sensitive to moisture, especially after heat treatment. Upon heating a vacuum is created inside the heating element, which sucks air inside. This makes control of ambient air humidity an important factor, especially after heat treatment. Moisture penetration depth analysis could show that the main reason of bad insulation is located close to the ends of the heating elements. Finally, the sealing of heating elements using UV curable adhesives made it possible to lower the element fabrication time by reducing the time of curing but improvements in temperature durability is necessary before can be used in large scale production.

**Keywords:** Heating elements, sealing, moisture, humidity, magnesium oxide (MgO), hydration, magnesium hydroxide ( $\text{Mg(OH)}_2$ ), insulation, electric conductivity, electric resistivity.

## Acknowledgements

This project was carried out between October 2018 and February 2019, at the company Backer AB and the division of Materials Engineering at LTH, Lund University.

I would like to thank my supervisor at LTH Dmytro Orlov for his great support and involvement in the project. He has always been willing to discuss different matters with me regarding the project and provided useful feedback during the progress. I would like to thank my supervisor at Backer Hans-Göran Künkel for his enthusiastic approach and his involvement in our many hours of discussion. I would also like to thank my examiner Srinivasan Iyengar who has always been inspiring and increased my knowledge in materials science with interesting discussions and lectures.

I would also like to thank to Johan Person Medin for making this project possible and sharing his technical knowledge. I would like to thank Pierre Kronvall for his involvement with providing samples but also sharing his network of colleagues within the company.

Many thanks to Backer for the opportunity to do my master thesis at the company and appreciations goes to all employees who has always been helpful, supportive and overall creating a nice environment to work in.

At last I would also like to thank my family for helping me in different ways to stay focused and who with love and care has always been a great support.

## Glossary with abbreviations

### **Adhesion**

The action or process of adhering to a surface or object.

### **Adhesives**

Substance used for sticking objects or materials together.

### **Annealing**

Heat treatment that can alter the physical properties of a material to increase ductility and reduce hardness.

### **Bolt**

Part of the element which connect the resistance wire to the application.

### **Climate chamber (CC)**

An environmental chamber used to test the effects of specific environmental conditions.

### **Electric conductivity**

A measure of a material's ability to conduct an electric current.

### **Dry stock (DS)**

Storage compartment where the humidity is lower relative to the atmosphere.

### **Epoxy**

Any type of resin or adhesive consisting of polymers of epoxides.

### **Humidity**

A quantity representing the amount of water vapor in the atmosphere or in a gas.

### **Hydration**

The process of combining a substance with water molecules.



### Hygroscopic

A material's or substance's ability to absorb moisture from ambient environment.

### Electric insulation

Action of preventing an electric current to travel through a material.

### Insulation pearl

Part of the element, used to increase electrical creeping distance to prevent current leakage.

### Magnesium oxide powder

White hygroscopic powder of used for electrical insulation due to its low electric conductivity.

### Magnesium hydroxide

Chemical compound  $Mg(OH)_2$  formed from hydration of magnesium oxide powder.

### Mantle

Metallic tube that surrounds the element and is in contact with the surroundings.

### Moisture

Water diffused in small quantities as vapor either within a solid or condensed on a surface.

### Polyurethane (PU)

Synthetic resin in which the polymer units are linked by urethane groups.

### Resin

Solid or liquid substance used as the basis for adhesives.

### Electric resistivity

Measure of the resisting power of a specific material to the flow of an electric current.

### Resistance wire

Core part of the element, which transfers heat from an electric current.

### Room temperature vulcanizing silicone (RTV)

RTV is a silicone based substance made from a two-component system, base material and curative.

### Sealing

Material or process used to prevent penetration of moisture and provides protection from ambient substances.

### Sealing ability

The impermeability of a material to prevent passage of ambient substances.

### UV curing

Process where ultraviolet light is used to initiate photochemical reaction that generates crosslinking of polymers.

# List of figures

*Figure 1: Diagram showing the correlation between absolute humidity, relative humidity and temperature. Red markers indicate environmental conditions used in the experiments. .... 4*

*Figure 2: Image of a resistance wire to a tubular heating element. .... 5*

*Figure 3: Image of the rolling of the mantle to tubular heating elements from metal strips. .... 6*

*Figure 4: Image of the filling of tubular heating elements (left), the rolling machines (center) and an X-ray image of the end of a tubular heating element after rolling (right). .... 7*

*Figure 5: Image of the annealing furnace where several tubular heating elements are about to enter. .... 8*

*Figure 6: Bending of a tubular heating element (left) and brazing (right) .... 9*

*Figure 7: Image of the sealing process for a heating element where several tubes are connected by a flange... 10*

*Figure 8: Schematic of a tubular heating element end. .... 11*

*Figure 9: Optical Image of a crack in the sealing of a tubular heating element. .... 12*

*Figure 10: Image where the adhesion of the sealing has been bad ..... 13*

*Figure 11: Photograph of CTS climate chamber used in the present investigation. .... 17*

*Figure 12: Image of FLUKE 1507 insulation tester used for measuring electrical resistance in tubular heating elements during testing..... 19*

*Figure 13: Image of the UV kit and the handheld UV light source (APM, UV-cure, LED 365 nm) procured from APM Technica..... 23*

*Figure 14: Evolution of electrical resistance of insulation material in annealed tubular heating element sample during exposure to moist air in dry stock (25 °C, 30 % humidity)..... 26*

*Figure 15: Evolution of electrical resistance of insulation material in annealed tubular heating element samples during exposure to moist air in climate chamber (25 °C, 90 % humidity)..... 27*

*Figure 16: Evolution of electrical resistance of insulation material in annealed tubular heating element samples during exposure to hot and moist air in climate chamber (70 °C, 90 % humidity). ..... 28*

*Figure 17: Evolution of electrical resistance of insulation material in annealed tubular heating element samples during the first 2 hours of exposure to moist air in ambient environment. .... 34*

*Figure 18: Evolution of electrical resistance of insulation material in annealed tubular heating element samples after pressure stabilization during exposure to moist air in dry stock (25 °C, 30 % humidity). ..... 36*

*Figure 19: Comparison of evolution of electrical resistance of insulation material in annealed tubular heating element samples during exposure to moist air in climate chamber (25 °C and 70 °C, 90 % humidity). ..... 38*

*Figure 20: X-ray image of a heating element with a prolonged bolt and no sealing where MgO has been removed in order to determine depth of moisture penetration..... 41*

*Figure 21: Evolution of relative decrease in electrical resistance of insulation material in annealed tubular heating element samples during the initial 30 minutes of exposure to moist air in ambient environment. .... 43*

*Figure 22: Design of a sealing utilizing both a UV-cured resin and RTV for attaching the insulation pearl..... 46*

## List of tables

<i>Table 1: Electrical conductivities and respective electrical resistivities for H<sub>2</sub>O, MgO and Mg(OH)<sub>2</sub> [7-9].</i>	<i>2</i>
<i>Table 2: Absolute humidity at different temperatures and relative humidity typical for the production environment at Backer.</i>	<i>3</i>
<i>Table 3: Conditions of exposure to moist air for the samples of tubular heating elements before annealing.</i>	<i>18</i>
<i>Table 4: Conditions of exposure to moist air for the samples of tubular heating elements. Initial resistance immediately after the exposure in CC is indicated in the right-hand side column.</i>	<i>18</i>
<i>Table 5: Conditions of exposure to moist air for the samples of tubular heating elements with extended bolt.</i>	<i>20</i>
<i>Table 6: Sealing material and storage time between annealing and sealing.</i>	<i>21</i>
<i>Table 7: Conditions of exposure to moist air for the samples of tubular heating elements with varying sealing materials inside the climate chamber.</i>	<i>22</i>
<i>Table 8: Critical parameters of UV adhesives selected for evaluation in this project.</i>	<i>23</i>
<i>Table 9: Resistance of insulation material in samples immediately after storage (before annealing).</i>	<i>25</i>
<i>Table 10: Effect of exposure conditions on the length of insulation material necessary to remove from the ends of a tubular heating element for restoring the level of electrical resistance to an acceptable level above the threshold value of 10 GΩ.</i>	<i>29</i>
<i>Table 11: Effect of exposure conditions on electrical resistance and resistance to short-circuit in high voltage (1600 V) in a tubular heating element sealed with PU.</i>	<i>30</i>
<i>Table 12: Effect of exposure conditions on electrical resistance and resistance to short-circuit in high voltage (1600 V) in a tubular heating element sealed with Araldite D.</i>	<i>30</i>
<i>Table 13: Effect of exposure conditions on electrical resistance and resistance to short-circuit in high voltage (1600 V) in a tubular heating element sealed with Ecobond.</i>	<i>31</i>

<i>Table 14: Effect of exposure conditions on electrical resistance and resistance to short-circuit in high voltage (1600 V) in a tubular heating element sealed with UV-curable adhesives. ....</i>	<i>32</i>
<i>Table 15: Resistance of insulation material in samples after exposure to ambient air in climate chamber and after 72 hours of storage in dry stock (25 °C, 30 % humidity).....</i>	<i>39</i>
<i>Table 16: Measured values from evolution of electrical resistance of insulation material in annealed tubular heating element samples during exposure to moist air in dry stock (25 °C, 30 % humidity). ....</i>	<i>54</i>
<i>Table 17: Measured values from evolution of electrical resistance of insulation material in annealed tubular heating element samples after exposure to moist air inside climate chamber for 24 hours at 25 °C in 90 % humidity. ....</i>	<i>56</i>
<i>Table 18: Measured values from evolution of electrical resistance of insulation material in annealed tubular heating element samples after exposure to moist air inside climate chamber for 72 hours at 25 °C in 90 % humidity. ....</i>	<i>58</i>
<i>Table 19: Measured values from evolution of electrical resistance of insulation material in annealed tubular heating element samples after exposure to moist air inside climate chamber for 24 hours at 70 °C in 90 % humidity. ....</i>	<i>60</i>
<i>Table 20: Measured values from studying of effect of exposure conditions on the length of insulation material necessary to remove from the ends of a tubular heating element for restoring the level of electrical resistance to an acceptable level above the threshold value of 10 GΩ. ....</i>	<i>62</i>
<i>Table 21: Measured values from studying of effect of exposure conditions on the length of insulation material necessary to remove from the ends of a tubular heating element for restoring the level of electrical resistance to an acceptable level above the threshold value of 10 GΩ after storage for 24, 48 and 72 hours (25 °C, 30 % humidity). ....</i>	<i>63</i>

*Table 22: Measured values from studying of effect of exposure conditions on the length of insulation material necessary to remove from the ends of a tubular heating element for restoring the level of electrical resistance to an acceptable level above the threshold value of 10 GΩ after storage for 72 hours (25 °C, 30 % humidity) and 72 hours (25 °C, 90 % humidity). ..... 64*

*Table 23: Measured values from studying of effect of exposure conditions on the length of insulation material necessary to remove from the ends of a tubular heating element for restoring the level of electrical resistance to an acceptable level above the threshold value of 10 GΩ after storage for 24 hours (70 °C, 90 % humidity). ..... 65*

*Table 24: Measured values from studying of effect of exposure conditions on the length of insulation material necessary to remove from the ends of a tubular heating element for restoring the level of electrical resistance to an acceptable level above the threshold value of 10 GΩ after storage for 48 hours (70 °C, 90 % humidity). ..... 66*

# Contents

Populärvetenskaplig sammanfattning .....	i
Popular science summary .....	ii
Abstrakt.....	iii
Abstract.....	iv
Acknowledgements .....	v
Glossary with abbreviations .....	vi
List of figures .....	ix
List of tables .....	xi
Contents .....	xiv
1. Introduction.....	1
1.1 Background .....	1
1.1.1 Electric resistivity as the measure of insulator efficiency.....	2
1.1.2 Humidity and the content of water in ambient environments.....	3
1.2 Production of heating elements .....	5
1.3 Properties of a sealing.....	12
1.3.1 Adhesion and sealing ability .....	13
1.3.2 High temperature durability .....	13
1.3.3 Curing.....	14
1.4 Novel materials for sealing .....	15
1.4.1 Glass ceramics .....	15
1.4.2 Polymers .....	15
1.5 Novel techniques of sealing.....	16
1.5.1 UV-curing .....	16
1.5.2 Film spraying .....	16
1.6 Objectives for the project.....	16
2. Materials and Methods .....	17
2.1 Sample preparation.....	17



2.2 Conditions of exposure to moist air and resistivity measurements .....	18
2.3 Moisture penetration test .....	20
2.4 Time effect on sealing .....	21
2.5 UV curing .....	23
3. Results .....	25
3.1 Dependence of electrical insulation on ambient environment .....	25
3.2 Hydration of MgO insulation from the moisture in ambient air.....	29
3.3 Effect of sealing material and exposure time .....	30
3.4 Testing of novel UV-curable resins.....	32
4. Discussion .....	33
4.1 Importance of insulation in tubular heating elements.....	33
4.2 Effect of hydration of MgO insulation from the moisture in ambient air .....	41
4.3 Effect of sealing material and exposure time .....	43
4.4 Improvement and challenges after testing of novel UV-curable resins.....	45
4.5 Additional comments .....	47
5. Conclusions.....	48
6. Future work .....	49
References.....	50
Appendices .....	54
Appendix I Changes in electrical insulation during exposure to ambient environment.....	54
Appendix II Hydration of MgO insulation from the moisture in ambient air .....	62

# 1. Introduction

## 1.1 Background

NIBE industries is a Swedish company, founded 1952 by Nils Bernerup, in Markaryd Sweden. Today NIBE is a global company with more than 15000 employees. NIBE was introduced in the stock market 1997 and 2018 had a turnover of almost 20 billion SEK [1].

The company has three main business areas: NIBE Climate Solutions, NIBE Stoves and NIBE Elements. NIBE Climate Solutions produces energy efficient solutions for household applications while NIBE Stoves produces stoves and fire places where design and energy efficiency is of main importance. The business area NIBE Element produces components for intelligent heating and steering [1].

NIBE Element consist of 57 sites in 21 countries with more than 8500 employees. The global head quarter of the business area NIBE Element today is Backer AB. Backer was founded by Christian Backer in Sösdala in 1949, which was later bought by NIBE. The site in Sösdala has a factory with approximately 250 employees of whom 70 are engineering personnel. The majority of production in Sösdala involves manufacturing of heating elements where the tubular heating element is one of the most common types [2].

Today tubular heating elements can be found almost anywhere from coffee machines to cab heaters. The ability to efficiently transfer heat from an electric current makes heating elements an efficient source for heating. The factory in Sösdala produces tubular heating elements in tens of millions on a yearly basis. The technology itself is used today as a modern way of intelligent heating, but many challenges are ahead.

Heating elements are very sensitive to moisture, and thus sealing is a necessity protecting the elements from such. Today the process of sealing takes place in latter stages of production, which increases the risk of moisture penetration of the heating elements. The sealing process itself is also not without problems. Many materials used for sealing require long curing time, which puts additional demand on an effective production. Separate compartments for sealing, health issues and insufficient curing are other aspects, faced as major challenges in today's production.

There is a major interest at Backer AB to evaluate and to improve production as well as to optimize the sealing of tubular heating elements.

### 1.1.1 Electric resistivity as the measure of insulator efficiency

Electrical insulation can be evaluated through the measurement of either electric resistivity or electric conductivity. Electrical resistivity is material resistance towards the flow of an electric current while electrical conductivity is the ability to conduct an electric current. Electrical resistivity ( $\rho$ ) is measured as Ohm meter ( $\Omega \cdot m$ ) while electrical conductivity ( $\sigma$ ) is measured as Siemens per meter (S/m) [3, 4].

Resistivity can be calculated using the following equation:

$\rho = R \cdot \frac{A}{L}$ , where R is electrical resistance, while A and L are the cross-section area and the length of a specimen, respectively.

Electrical conductivity is the reciprocal of electrical resistivity:

$$\sigma = \frac{1}{\rho}.$$

This correlation becomes important as resistance (R) is commonly measured when testing heating elements.

Magnesium oxide (MgO) is used as electrical insulating material inside the element. The low electrical conductivity of MgO makes it an excellent electrical insulator by preventing electrical leakage inside the heating element. This combined with good thermal conductivity makes MgO the material of choice for electrical insulation in heating elements [5, 6]. MgO is hygroscopic, which gives it the ability to attract and absorb water [7-9]. In the presence of water, magnesium oxide reacts and forms magnesium hydroxide (Mg(OH)<sub>2</sub>) in the following reaction: MgO(s) + H<sub>2</sub>O (g/l) → Mg(OH)<sub>2</sub>(s), where s, g and l stand for solid, gaseous and liquid states of respective reactants [5, 8]. Mg(OH)<sub>2</sub> is also hygroscopic and thus can also absorb water. When Mg(OH)<sub>2</sub> absorbs water (hydration) it binds water molecules along particle surfaces and in grain boundaries. This enables ion exchanges between the surface and the interior of the Mg(OH)<sub>2</sub> particles [10-12]. This has major influence on electrical insulation of the heating elements as the ion exchanges in Mg(OH)<sub>2</sub> particles increases electrical conductivity in the electric insulating material. The combination of hydrated Mg(OH)<sub>2</sub> and the presence of free water inside the heating elements is believed to have a major effect on the electric insulation properties. The reversed reaction of Mg(OH)<sub>2</sub> to MgO is possible but only at temperatures above 350 °C [7, 8, 10]. In Table 1, a summary of different electrical conductivities and corresponding resistivity ranges can be found for the materials of interest.

Table 1: Electrical conductivities and respective electrical resistivities for H<sub>2</sub>O, MgO and Mg(OH)<sub>2</sub> [7-9].

Material	Condition	Conductivity (S/m)	Resistivity ( $\Omega \cdot m$ )
H <sub>2</sub> O	Tap water	10 <sup>-2</sup> -10 <sup>-3</sup>	10 <sup>2</sup> -10 <sup>3</sup>
MgO	Dry	10 <sup>-16</sup> -10 <sup>-17</sup>	10 <sup>16</sup> -10 <sup>17</sup>
Mg(OH) <sub>2</sub>	Dry	10 <sup>-12</sup> -10 <sup>-13</sup>	10 <sup>12</sup> -10 <sup>13</sup>
Mg(OH) <sub>2</sub>	Wet (18 wt% water)	10 <sup>-3</sup> -10 <sup>-4</sup>	10 <sup>3</sup> -10 <sup>4</sup>

As can be seen in Table 1 the presence of moisture has a major influence on electrical conductivity. This proves the necessity of a sealing in order to prevent moisture from entering the heating elements.

### 1.1.2 Humidity and the content of water in ambient environments

The source of moisture during the production of heating elements is the ambient air, and thus the understanding of air humidity becomes important. Humidity can be described as the amount of water present in the air. It is measured as either absolute humidity (g H<sub>2</sub>O/m<sup>3</sup> air) or as relative humidity (%) [13]. The latter is often found in flow meters showing the humidity of controlled environments. In Table 2, water content (absolute humidity) can be seen at different temperatures and relative humidity.

*Table 2: Absolute humidity at different temperatures and relative humidity typical for the production environment at Backer.*

<b>Temperature (°C)</b>	<b>Relative humidity (%)</b>	<b>Absolute humidity (g water/kg air)</b>
-20	50	0,6
-20	100	1,21
-10	50	1,13
-10	100	2,25
0	50	2,1
0	100	4,2
10	50	3,92
10	100	7,84
20	50	7,31
20	100	14,62
30	50	13,63
30	100	27,27

When temperature is constant, the water content increases linearly with increase in relative humidity. The dependence is different if the relative humidity is kept constant while the temperature increases. The reason for this is that relative humidity is measured as the ratio of partial pressure of water vapor in the mixture of pure vapor pressure of water at a certain temperature. This means that in order to determine the content of water (absolute humidity) both temperature and relative humidity need to be known [13]. The calculated diagram of absolute humidity is shown for a temperature range of 0-100 °C and relative humidity range of 0-100 %.

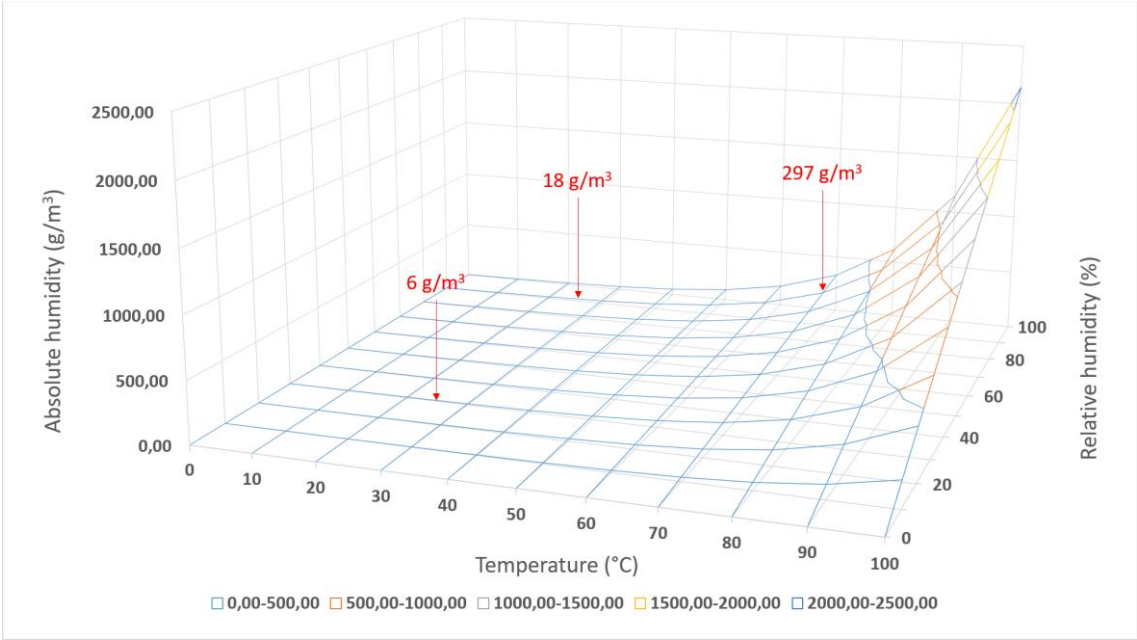


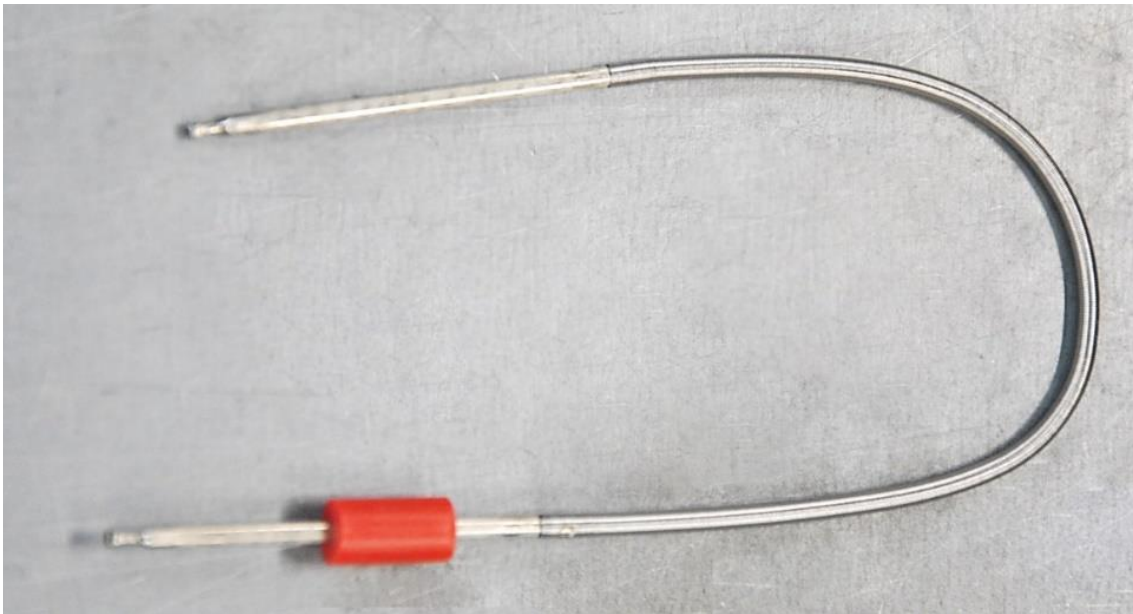
Figure 1: Diagram showing the correlation between absolute humidity, relative humidity and temperature. Red markers indicate environmental conditions used in the experiments.

It becomes clear from Figure 1 that the content of water (absolute humidity) is much higher at elevated temperatures. This makes year season an important factor in the production of heating elements. During winter, ambient temperature is low and thus humidity becomes less of a problem since ambient air does not contain as much water, even at high relative humidity. During summer, ambient temperature increases making it possible for air to have a higher water content. This increases the presence of water making it harder to prevent moisture penetration of heating elements during production.

## 1.2 Production of heating elements

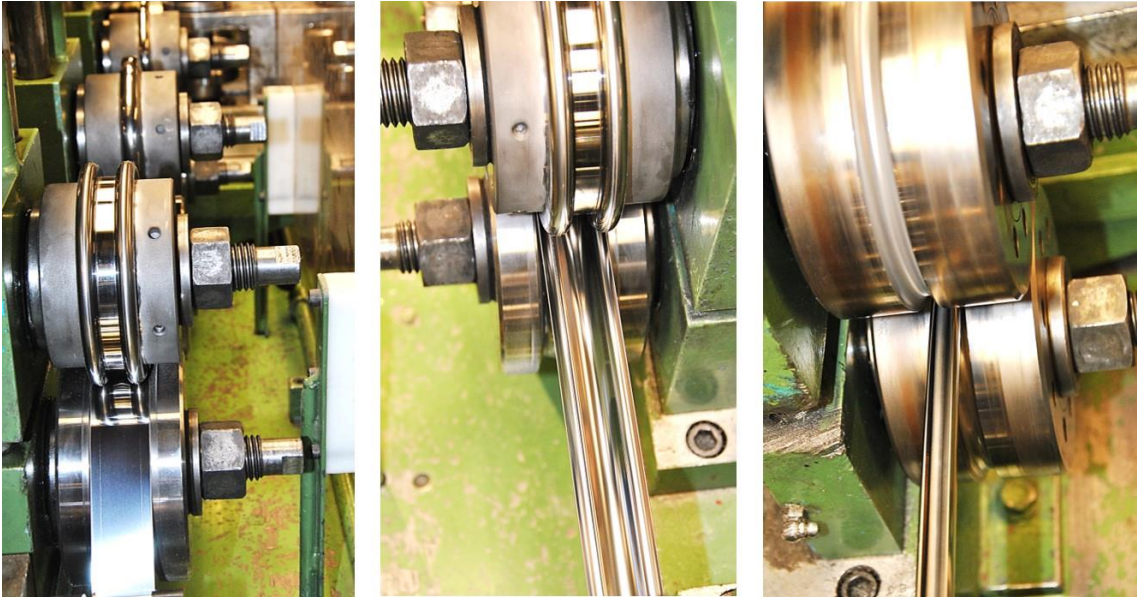
Heating elements are produced in different designs, where 'tubular heating elements' is one of the most common types.

The production of tubular heating elements starts with the resistance wire. The resistance wire is an important part of the heating element as it transfers heat to the surroundings, when active. The resistance wire is placed in the core of the tubular heating elements and when active an electric current is conducted through it. The resistance wire then transfers heat from the electric current to the mantle, and through it to the surrounding medium of the heating element. The resistance wire is connected to bolts at each end of the wire, connected by spot welding. These bolts connects the heating element to external equipment and the electric source. An image of a resistance wire can be seen in Figure 2.



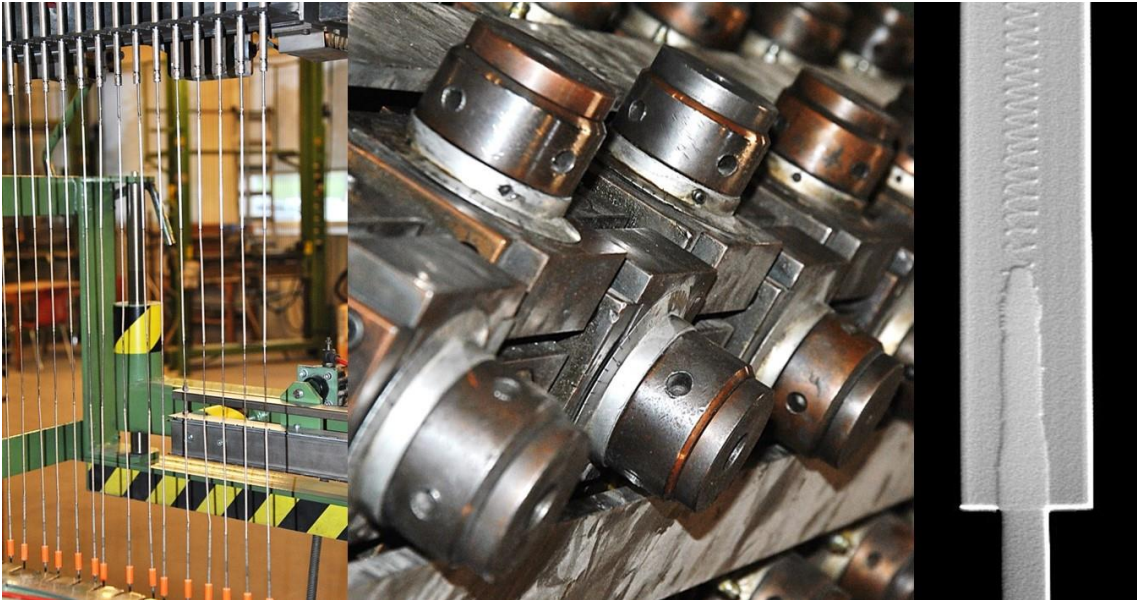
*Figure 2: Image of a resistance wire to a tubular heating element.*

The mantle of the tubular heating elements is produced from metal strips in stainless steel (SS304), which are shaped into a tube by rolling and welding. The rolling of metal strips into a tube can be seen in Figure 3.



*Figure 3: Image of the rolling of the mantle to tubular heating elements from metal strips.*

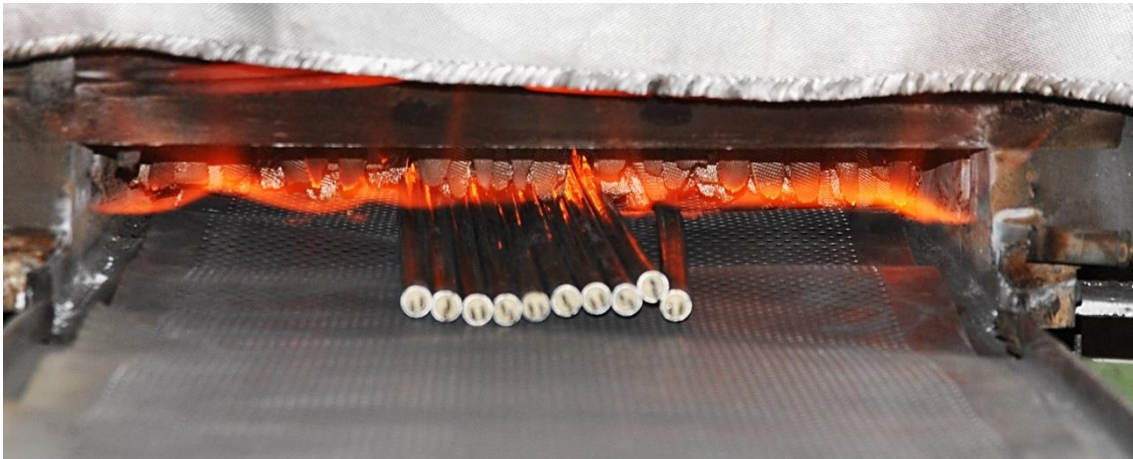
Next step in production is the filling of electric insulating material (MgO). The tubes are moved to filling machines where the resistance wire is connected to the mantle using a plastic brick. The tubes are filled with magnesium oxide powder (MgO), and during filling the machine stretches the resistance wire to its full length. Production then continues with the tubes being rolled, which compresses the tubes and the MgO powder is compacted into a porous solid. This ceramic-like structure becomes the electric insulator of a finished tubular heating element. Images from these steps in production can be seen in Figure 4.



*Figure 4: Image of the filling of tubular heating elements (left), the rolling machines (center) and an X-ray image of the end of a tubular heating element after rolling (right).*



After rolling, the tubes are annealed. The temperature inside the annealing furnace is very high (1080 °C), and the ambient atmosphere (inside the furnace) is free of oxygen. The annealing increases the ductility through stress relief of austenitic stainless steel, which enables bending of the tube (mantle) [14]. The high temperature also reverses the formation of  $Mg(OH)_2$  back to  $MgO$  [10] while also removing possible moisture, present from the filling process. The temperature increases the pressure inside the heating elements and hot air is forced out of the heating elements, inside the furnace. This creates a vacuum inside the heating elements when leaving the furnace. An image of tubular heating elements entering the annealing furnace can be seen in Figure 5.



*Figure 5: Image of the annealing furnace where several tubular heating elements are about to enter.*

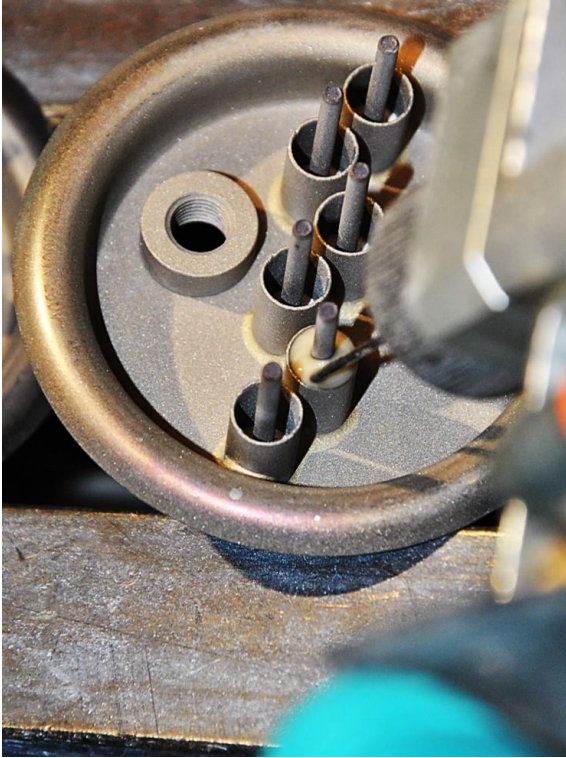
After annealing the heating elements need to cool down or they may crack when shaped otherwise. Once the heating elements has cooled, they are shaped according to the final application of the heating element. The production then continues with brazing and welding where flanges are attached to the heating elements. This makes the ends of the heating elements very hot and is one reason why the sealing takes place at latter stages in the production. The bending of a tubular heating element and brazing of a flange can be seen in Figure 6.



*Figure 6: Bending of a tubular heating element (left) and brazing (right)*

Before elements can be sealed, good electrical resistance must be obtained. This is achieved by drying operations at various temperatures in the production. The elements are sealed only when dry and a threshold of 10 GΩ is often used to ensure good electrical insulation to meet the requirements of final application. Different types of sealing materials are used, and thus the sealing process may vary depending on the sealing material used. For heating elements operating in air respective temperature is often high, and material such as room temperature vulcanizing silicone (RTV) is often used due to its high thermal durability. In some cases where temperature prevents the usage of RTV sometimes only a silicone plug is used. The reason for this is that the high temperature prevents usage of most sealing materials as they tend to fail due to bad adhesion to the heating element. However, in this case the temperature itself protects the heating element by acting as a protective barrier to moisture. This makes the low sealing ability of the silicone plug a minor issue but its high thermal durability makes it work as an efficient sealing in very hot environments.

For heating elements operating in water or liquid medium, respective temperature is lower, but the sealing ability and adhesion of materials becomes more important. Polyurethane (PU) and Epoxy-based materials are commonly used in these types of elements due to their great adhesion and sealing ability [15, 16]. For RTV, PU and Epoxy curing is required, but the time of curing varies significantly. For some sealing materials, curing process requires heat to initiate as well as to decrease curing time [17-19].



*Figure 7: Image of the sealing process for a heating element where several tubes are connected by a flange.*

At each end of a tubular heating element there is an insulation pearl. It is made of plastic or ceramic material and is attached to the heating element by the sealing, during curing. The main purpose of the insulation pearl is to limit the risk of current leakage by increasing the electric creeping distance between the bolt and the mantle of the heating element. The definition of current leakage may vary but at Backer it is commonly defined as when electric current is conducted between the bolt/ resistance wire to the mantle with a resistance below the threshold of 10 GΩ. After the sealing process is finished the heating elements are ready for transport. At this point, wires and electronics can also be connected to the heating elements by the bolts. A schematic of a tubular heating element end with the important parts mentioned in the Production of heating elements section can be viewed in Figure 8.

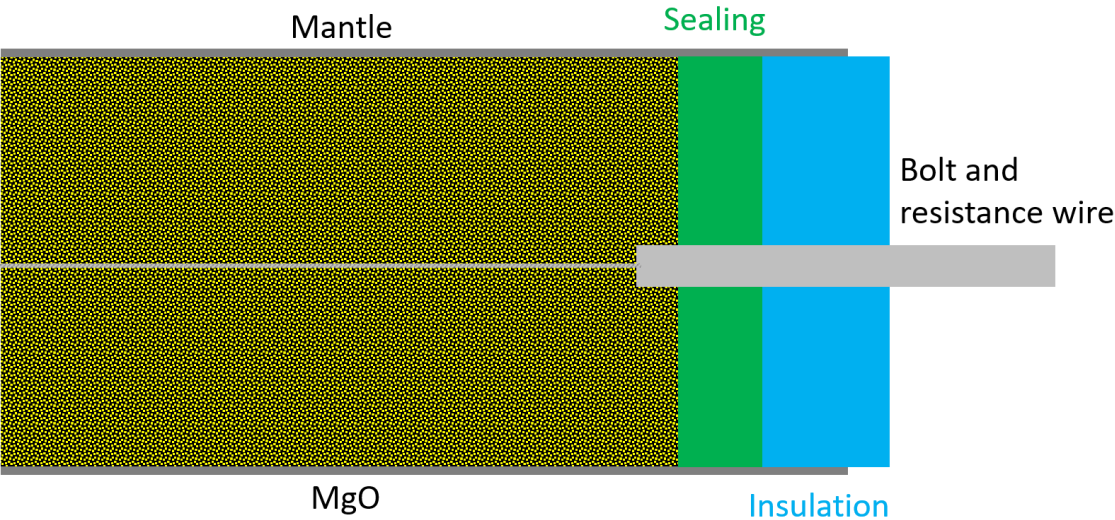
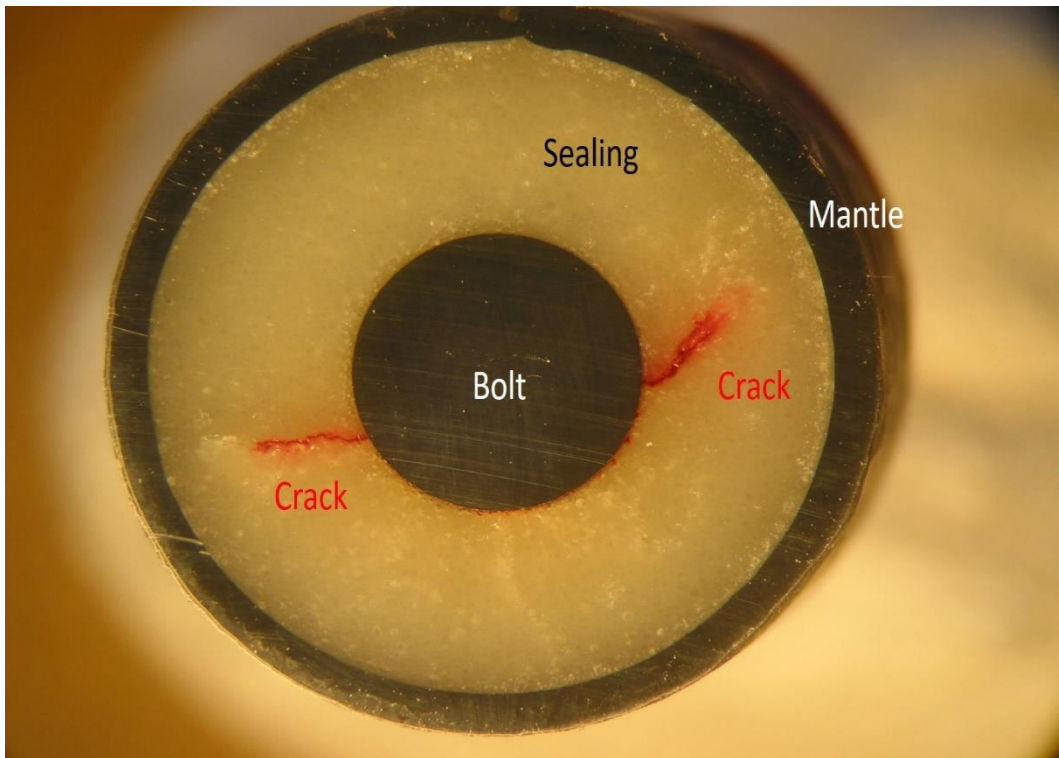


Figure 8: Schematic of a tubular heating element end.

### 1.3 Properties of a sealing

When taking into consideration all the aspects of tubular heating element production relevant to insulation and humidity, it becomes clear why sealing is necessary. Therefore, the sealing itself needs to fulfil different requirements in order to be used. Since the applications of heating elements may vary, properties have varying levels of importance depending on respective environments. In the automotive industry, the sealing needs to withstand vibration and sudden impacts, or otherwise cracks may form, see an example Figure 9. Properties such as adhesion, sealing ability and high temperature durability are other properties of great importance for sealing materials.



*Figure 9: Optical Image of a crack in the sealing of a tubular heating element.*



### 1.3.1 Adhesion and sealing ability

In some applications, especially for elements operating in water, the environment can become very humid. Elements operating in these conditions need both good adhesion and excellent sealing ability. The ability to wet a surface and form an interface between the mantle and the sealing makes is what gives a sealing material good adhesion [16, 20, 21]. Adhesion is also important in preventing channels between the mantle and the sealing. Such channels are often the reason of moisture penetration into the heating elements, after sealing [16, 22]. Since heating elements operate in cycles where they become warm and cold, metallic components expand as temperature increases. When this happens, good adhesion must ensure that sealing remains in contact with metal. Sealing ability can be described as the ability to prevent moisture from traveling through the material interface [15, 16, 20, 23]. It is important since it limits the possibility of forming channels inside the sealing material thus limiting moisture penetration into the element. In Figure 10, the formation of channels along the bolt part enabled moisture (red) to penetrate through the sealing. This was the result of bad adhesion and/or bad sealing ability [15, 16].

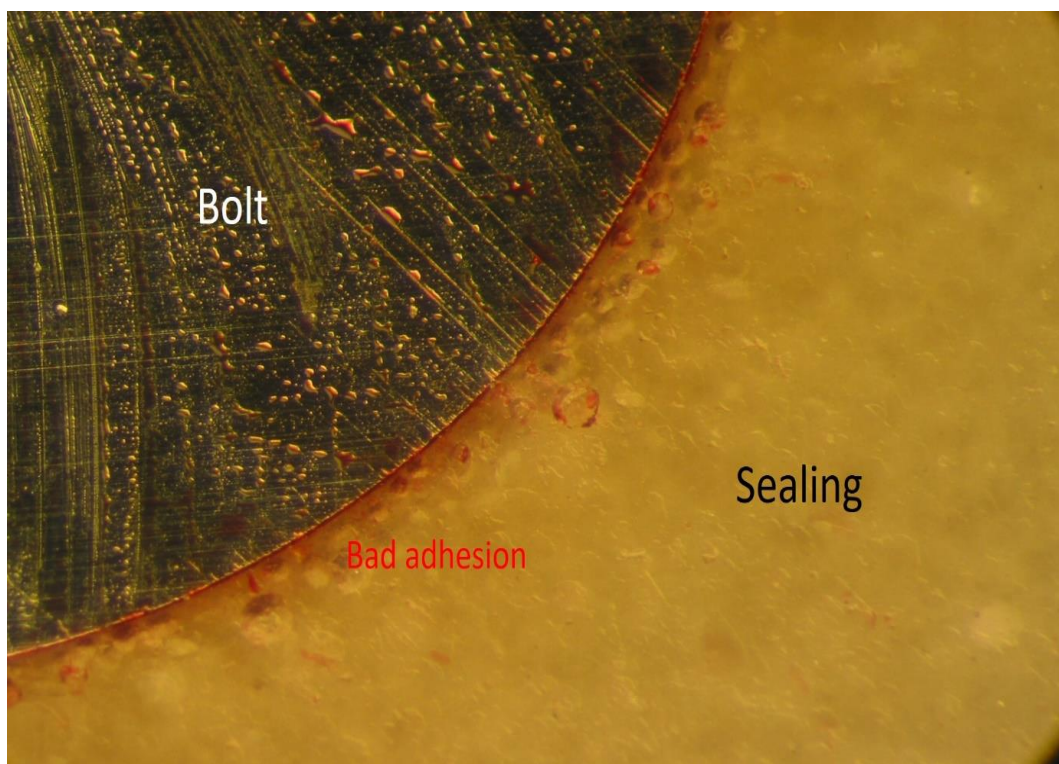


Figure 10: Image where the adhesion of the sealing has been bad

### 1.3.2 High temperature durability

In high temperature applications, such as elements operating in air, the importance of sealing ability decreases. The temperature itself acts as a protective barrier against moisture when active, and thus it becomes more important that the sealing remains durable at elevated temperatures. The ability to not deform and stick to the heating element becomes more important when the operating temperature of environment increases [24, 25]. Because of this, sealing materials used in such applications are typically not good in preventing moisture penetration into the element when inactive. This becomes a problem during transportation and storage for longer periods. In order to prevent moisture penetration during such time periods, a thin layer of silicone oil can be added sometimes. This burns off when an element is active but protects the element until then [26, 27].

### 1.3.3 Curing

In many cases, sealing material is applied as a liquid, which needs to cure before it starts working as an efficient sealing. When applied as liquid, it is important that sufficient time is given for the sealing material to fill out deficiencies in the mantle and on the surface of the MgO, in order to obtain good adhesion [15-17, 19]. This sometimes become a problem if the material has high viscosity since this will decrease the flow ability of the material. Higher also viscosity increases the risk of air bubbles, forming in the sealing. The formation of air bubbles makes the sealing less impermeable, which decreases the sealing ability of the material. This formation of air bubbles can also become an issue if temperature is too high when the sealing is applied. If so, the liquid might boil and if cured before these bubbles are gone they can become trapped inside the sealing [15, 16, 22]. The time of curing is another important aspect. Curing time can vary significantly depending on the material and method used, and in some cases may take up to 24 hours or more. Curing can often be influenced by temperature, as this helps to accelerate the curing process, often used for thermal curing techniques [16, 17, 19]. In some cases, elevated temperature is also a requirement for the curing to initiate as the initiator of polymerization is only activated at elevated temperatures, which is the case for many Epoxy-based sealing materials. Several types of curing are available at present day but the most common technique is thermal curing. In recent years, the option of UV curing has become more popular, but it is mostly found within electronics industry [19, 28-31].

## 1.4 Novel materials for sealing

There are many different studies and industries from which knowledge can be transferred for improving the sealing of heating elements. Glass ceramics and different polymers are materials with a potential to work as efficient sealing materials while UV curing and film spraying are methods that could be used for improving the sealing process.

### 1.4.1 Glass ceramics

Glass-ceramics combine the properties of crystalline ceramics and the ease of processing of glass [20, 21, 32]. They possess great adhesion and bond effectively with metals and alloys. When applying a glass-ceramic as a sealing, it needs to be melted first. The melt can flow into cavities on the metal surface, wetting the surface before creating an interface between the metal and the glass ceramic. This combined with the ability to affect thermal expansion potentially makes glass-ceramics a good sealing material when working with metals and alloys [20, 21, 32]. They also possess excellent high-temperature durability. The combination of adhesion to metal and high temperature stability is not commonly found among sealing materials used today, which should make glass-ceramics a suitable sealing material in high-temperature applications [20, 21, 25, 32, 33].

Even though glass-ceramics shows promise as a sealing material, there are still many challenges before they can be used for large scale production. They require high melting temperature, which makes them difficult to handle as well as putting high demands on equipment used. They are also more brittle compared to plastic materials or polymers, which can become an issue if used in automotive industry where vibrations and impacts may deform or break the sealing.

### 1.4.2 Polymers

Polymers are widely used today for coating in different industries where coating of PCBs in electronic industry is one of the more common application fields [24, 26, 34-36]. By choosing correct monomer, several different properties can be achieved. Today, Backer AB uses both PU and Epoxy as sealing materials, which after curing works as efficient sealing in heating elements. [19, 29]. They possess excellent adhesion to metals and steels along with very good sealing ability. They also have outstanding electrical insulation, good thermal durability and high tensile strength. Furthermore, they are resistant to chemical reagents and corrosion, which makes them ideal sealing materials in heating elements [19, 23, 29-31, 37]. A problem with these materials is that they can be absorbed on respiratory ways or in contact with skin, from which allergic reaction may develop. This makes it difficult when working with PU and Epoxy based sealing materials as they need to be handled with care in controlled environments [19, 30, 31]. Today, there is a major interest at Backer in finding sealing materials with similar properties to both PU and Epoxy but without the health-related problems.



## 1.5 Novel techniques of sealing

Novel sealing materials require novel techniques for applying. Those promising in the context of applications for tubular heating elements are presented below.

### 1.5.1 UV-curing

A problem today with the curing of many sealing materials is the requirement of significant time and thermal energy (thermal curing). Recent studies have shown promise in replacing thermal curing with UV curing. The latter utilizes ultra-violet radiation as an energy source to initiate the curing process through radical polymerization. It has fast reaction kinetics, is less energy consuming and is applicable to different types of coatings and adhesives [17-19, 25, 29-31, 38]. The main components of UV-curable materials are: monomer, photo initiator, reactive diluent and additives. The photo initiator enables UV-curing process to initiate by working as an active site for polymerization. The monomer is the main building block while the additives mainly affect properties of the material. Materials such as epoxy resins have been successfully cured using a UV-radiation source with a potential to reduce curing time and energy consumption in comparison to thermal curing [19, 29-31, 37]. However, there are limitations with UV-curing such as the depth of light penetration. Since the source of energy is UV light, it is important that large area of the material is exposed to UV light source. This may become a problem when working with small surface to volume ratios. The UV-curing method is therefore well applicable for coatings or thinner types of sealing but might be more difficult to utilize when the sealing is of a thicker kind [15, 16, 22]. With the intention to solve this problem, studies on combining a photo initiator with a thermal initiator have been carried out. Most polymerizations are exothermic, which means that heat is released when the polymerization starts [29, 31, 37]. These studies showed that UV-light can trigger the initiation of a polymerization from which heat is released, and this triggers continuous thermal polymerization during the curing process. This combines the effectiveness of both UV- and thermal curing processes.

### 1.5.2 Film spraying

Today, sealing materials are typically applied in a liquid state. In electronic industry new ways of coating printed circuit boards (PCB) have been in development in recent years. Film spraying is a method where a thin film is sprayed onto the PCB and then allowed to cure [27, 39]. This method requires small quantity of material since the film is very thin, which also enables fast curing. Silicone based materials have been used successfully to protect PCBs from corrosion using this method. A similar method may provide a new way of applying sealing to the elements. The advantages of this method include the efficient utilization of sealing material and the ease of automation [34, 40, 41]. However, concerns regarding using this method for thicker coatings still remain since fast curing can be associated only with the small amount of material used. Such materials also do not need to withstand elevated temperatures, which is the case for many heating elements.

## 1.6 Objectives for the project

1. Creating a platform for continuous improving the sealing of heating elements.
2. Investigating the effect of exposure to humid environments on electrical resistivity of insulating material within heating elements.
3. Examining different materials and techniques for the sealing of heating elements.

## 2. Materials and Methods

Experimental work carried out in this project include preparation of samples for the investigation, selecting representative environmental conditions for the storage of heating elements, measurement of resistance in insulating materials, and testing of curing in new sealing materials. Samples prepared for the investigations included tubular heating elements annealed and then stored for various durations in controlled environments. Electrical resistance was continuously measured during storage for the following analysis of electrical insulation evolution over time. The testing of curing involved the measurement of sealing ability of procured adhesives along with electrical resistance evolution over time.

### 2.1 Sample preparation

The preparation of samples for the investigations involved the shaping of stainless steel SS304 strips into tubes. The dimension of the tubes were 550-600 mm in length and 8.5 mm in diameter after rolling. The tubes were then filled with MgO powder and rolled according to standard technological process at Backer AB. Afterwards, silicone plugs were attached to each end of the tube in order to prevent the loss of MgO powder and to limit moisture penetration during storage.

Prepared samples were stored inside a “dry stock” (DS) compartment having a temperature of 25 °C and a humidity of 30 %. The levels of temperature and humidity inside DS were allowed to vary within  $\pm 2$  °C and  $\pm 5$  %, respectively. For the process of annealing, silicone plugs were removed from the samples, and immediately afterwards the latter were placed either in the dry stock or in a climate chamber. Annealing was carried out at 1080 °C in inert atmosphere with no oxygen. The climate chamber (Clima Temperatur Systeme) used in this work is presented in Figure 11.



*Figure 11: Photograph of CTS climate chamber used in the present investigation.*

## 2.2 Conditions of exposure to moist air and resistivity measurements

In the first set of testing, an exposure to moist air, samples were stored in the climate chamber before annealing. Various conditions were used inside the climate chamber in order to control the concentrations of water vapor in ambient air. The storage conditions inside the climate chamber are summarized in Table 3. After storage electrical resistance was measured for the samples inside a dry stock.

*Table 3: Conditions of exposure to moist air for the samples of tubular heating elements before annealing.*

Sample	Time stored (h)	Temperature (°C)	Humidity (%)	Silicone plug removed
1	72	25	90	No
2				Yes
3			30	No
4				Yes

After the initial measurement of electric resistance, the samples were annealed at 1080 °C in a controlled atmosphere with no oxygen. After annealing the samples were returned inside the dry stock and electrical resistance was measured again.

In a second exposure test, samples were placed in the climate chamber or directly in the dry stock, after annealing. The varying storage conditions for the samples along with the environment inside the climate chamber are summarized in Table 4.

*Table 4: Conditions of exposure to moist air for the samples of tubular heating elements. Initial resistance immediately after the exposure in CC is indicated in the right-hand side column.*

Time stored in CC (h)	Temperature (°C)	Humidity (%)	Initial resistance (MΩ)
-	25	30	$3 \times 10^7$
24		90	11,3
72			3,5
24	70	90	4,4
72			0,5

After the moist air exposure inside the climate chamber, the samples were marked and placed inside the dry stock at 25 °C with 30 % relative humidity (6 g H<sub>2</sub>O/m<sup>3</sup> air). Electrical resistance was then measured every hour during the first day followed by measurement on a daily basis for the rest of the experiment. All measurements were carried out inside the dry stock. Electrical resistance was measured using a mobile FLUKE 1507 insulation tester, see Figure 12, which made it possible to perform measurements inside the dry stock. In order to judge the quality of the heating element electrical insulation, a threshold value of 10 GΩ was used, which is a standard used by Backer AB. The insulation tester unit had an upper limit of measurement at 11 GΩ, which was sufficient for the evaluation using the above criterion.



*Figure 12: Image of FLUKE 1507 insulation tester used for measuring electrical resistance in tubular heating elements during testing.*

The measurements were carried out by connecting two contact wires from the insulation tester unit to the sample. One wire was connected to the bolt while the other one to the mantle of the tube. A voltage of 1000 V was applied, from which a resistance was given. In order to carry out electrical measurements of resistance above 11 GΩ, a stationary insulation tester instrument was used. This instrument was stationary and therefore not possible to move into the dry stock. Due to this, the instrument was only used to evaluate the initial values of electric resistance and in excess of 11 GΩ.

### 2.3 Moisture penetration test

In order to investigate the presence of moisture inside the heating elements, a new set of samples was prepared using procedure similar to that described in section “2.1 Sample preparation” above. The only difference was an extended bolt in the new samples, which made it possible to remove sections of the mantle and the MgO solid while still being able to measure resistance. The samples were exposed to various moist air conditions during a storage inside the climate chamber before each measurement. The storage conditions for these samples are summarized in Table 5. After exposure, the samples were ground, and 20 mm sections of the mantle and MgO was removed from each end. Electrical resistance was then measured, and when necessary further removal of 20 mm sections of MgO and mantle was removed. This process was repeated until the resistance reached values above the threshold of 10 GΩ. An image of an analyzed sample can be seen in section 4.2 Effect of hydration of MgO insulation from the moisture in ambient air, see Figure 20.

*Table 5: Conditions of exposure to moist air for the samples of tubular heating elements with extended bolt.*

<b>Time stored (h)</b>	<b>Temperature (°C)</b>	<b>Humidity (%)</b>
24	25	30
48		
72		
24		90
48		
72		

## 2.4 Time effect on sealing

In order to evaluate time necessary before sealing heating elements, the samples were annealed and then U-shaped. Half of these samples were sealed immediately after being U-shaped while the other half were placed inside the dry stock for 72 hours. After 72 hours of storage inside the dry stock, the samples were dried out in a furnace at 200 °C for 12 hours, followed by cooling in a hot chamber at 70 °C until manageable by hand. Once manageable the samples were cleaned with compressed air to remove dust and loose MgO particles before being sealed. Three different sealing materials for testing were provided by the company for this experiment. After sealing, the samples were placed in the dry stock, and electrical resistance was measured on a daily basis for the duration of the experiment. The material and the time of sealing (after annealing) are summarized in Table 6.

*Table 6: Sealing material and storage time between annealing and sealing.*

<b>Sealing material</b>	<b>Time of sealing after annealing</b>	<b>Resistance after sealing (MΩ)</b>
PU 403	Immediately	≥11000
PU 403	After 72 hours	≥11000
Araldite D (epoxy)	Immediately	≥11000
Araldite D (epoxy)	After 72 hours	≥11000
Ecobond (epoxy)	Immediately	≥11000
Ecobond (epoxy)	After 72 hours	≥11000

To investigate further the effect of sealing time on sealing capacity, the samples were placed inside the climate chamber. After the exposure to moist air inside the climate chamber, the samples were returned to the dry stock and electrical resistance was measured again. The varying exposure conditions used inside the climate chamber are summarized in Table 7.

*Table 7: Conditions of exposure to moist air for the samples of tubular heating elements with varying sealing materials inside the climate chamber.*

<b>Time stored (h)</b>	<b>Temperature (°C)</b>	<b>Humidity (%)</b>
24	25	90
72	25	90
24	70	90
72	70	90

Furthermore, a high-voltage testing was carried out in order to investigate its effect on the sample performance. In this testing, a voltage of up to 1600 V was applied and signs of current leakage was measured instead of electrical resistance. The definition of current leakage may vary but in this test it was defined as a rapid increase of electric current capable of travelling from the resistance wire to the mantle when voltage was increased up to 1600 V.

## 2.5 UV curing

In order to investigate possible new sealing materials and methods, a mobile hand-held UV kit was procured from APM Technica. Figure 13 shows the UV kit and the handheld UV light source (APM, UV cure, 365 nm).



Figure 13: Image of the UV kit and the handheld UV light source (APM, UV-cure, LED 365 nm) procured from APM Technica.

Corresponding UV-curing adhesives were selected based on properties such as curing time, operating temperature range, viscosity and adhesion to metal. Respective parameters for the procured adhesives are summarized in Table 8.

Table 8: Critical parameters of UV adhesives selected for evaluation in this project.

Adhesive	Material	Curing type	Curing time	Temperature range	Viscosity ( $\text{kg m}^{-1} \text{s}^{-1}$ )	Additional comment
<b>Unocol 818</b>	Acrylate	UV	Seconds	-40°C, +110°C	0,7	1 component, good adhesion
<b>NOA 81</b>	Epoxy	UV	Seconds	-40°C, +110°C	0,3	1 component, great adhesion
<b>NEA 121</b>	Epoxy	UV	Seconds	-40°C, +110°C	0,3	1 component, great adhesion



The testing of the UV-curable adhesives was carried out on annealed and U-shaped samples. The samples were dried out at 200 °C for 12 hours followed by cooling inside a hot chamber at 70 °C. When manageable, the samples were cleaned using compressed air to remove dust and loose MgO powder in a similar fashion as mentioned in section 2.4 Time effect on sealing. Electrical resistance was then measured using the FLUKE 1507 insulation tester (see Figure 12) to check the sample quality. The adhesives were applied in liquid form in the amount sufficient to cover the surface of MgO insulation solid. Afterwards, curing was executed using the UV light source. Once cured, electrical resistance of the assembled sealed samples was measured again to make sure that the adhesive had cured but also to provide an initial electric insulation value for upcoming exposure testing. Finally, the samples were placed in the climate chamber in order to investigate the sealing capability of the adhesives in various environments. The exposure conditions used in the climate chamber are summarized in Table 7. After each storage inside the climate chamber, electrical resistance was measured repeatedly, and high voltage testing was carried out for each sample in a similar fashion as described in section 2.4 Time effect on sealing.

### 3. Results

The results of the electrical insulation measurements are presented as diagrams where the changes in resistance are shown over a period of time. Measurements on sealing ability, short circuit and moisture penetration of MgO are presented in tables.

#### 3.1 Dependence of electrical insulation on ambient environment

The resistance of insulation material in four samples that were stored in different conditions before annealing was measured after the storage in climate chamber. The same samples were the annealed after which resistance was measured once again. The measured resistance after storage in the climate chamber and after annealing are summarized in Table 9 and the storage conditions for each sample is presented in Table 3.

*Table 9: Resistance of insulation material in samples immediately after storage (before annealing).*

<b>Sample</b>	<b>Resistance after CC (MΩ)</b>	<b>Resistance after annealing (MΩ)</b>
<b>1</b>	67	$\geq 11000$
<b>2</b>	0	$\geq 11000$
<b>3</b>	627	$\geq 11000$
<b>4</b>	34	$\geq 11000$

The combination of low water content and lesser amount of MgO in direct contact with the air is what gives sample 3 the highest resistance. The opposite can be said about sample 2 where MgO was exposed more, and humidity was higher, thus resulting in the lowest resistance among the four samples. After annealing, all samples showed a high resistance, which proves that the annealing furnace effectively removes moisture from the heating elements.

The effect of exposure to air with a relatively low level of moisture is studied in the annealed tubular heating element samples by placing them in the dry stock at 25 °C and 30 % relative humidity (6 g H<sub>2</sub>O /m<sup>3</sup> air) immediately after annealing. Respective results for one of these samples are presented in Figure 14. The initial electrical resistance is very high (see Table 4) but drops very fast during the first hours of storage after annealing. Resistance then continues to decrease at a lower rate until 24 hours. Then, it increases slightly for a short period of time until 48 hours. After that, resistance decreases continuously for the rest of experiment but with small changes in electrical resistance. The variation of electrical resistance during this period can be associated with the variation of environmental conditions in the dry stock because of operational reasons at the factory. When looking at the measured values of resistance, it can be observed that it may increase overnight but then decreases during the following day. This behavior can be seen throughout the entire measurement with varying degree of clearness, see for instance the time period between 600 hours and 800 hours. After the initial drop, resistance remains below the threshold value of 10 GΩ throughout the entire measurement.

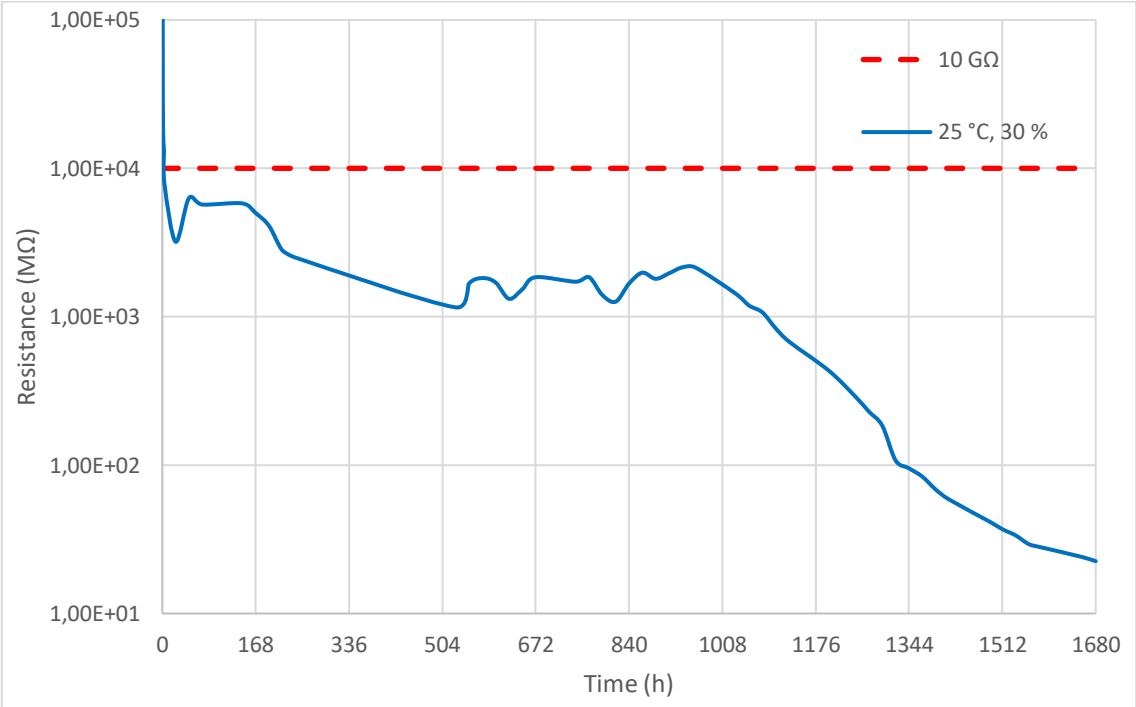


Figure 14: Evolution of electrical resistance of insulation material in annealed tubular heating element sample during exposure to moist air in dry stock (25 °C, 30 % humidity).

The effect of exposure to air with a relatively high level of moisture was studied in the annealed tubular heating element samples by placing them in the climate chamber for 24 hours at 25 °C in 90 % relative humidity (18 g H<sub>2</sub>O /m<sup>3</sup> air).

The electrical resistance decreases rapidly during exposure to humid air inside the climate chamber and the initial resistance value therefore is very low (see Table 4). When extracting heating element samples from the climate chamber and placing them inside the dry stock, electrical resistance is already below the threshold value of 10 GΩ. This shows that the decrease in electrical resistance of the material takes place so quickly that the first measurement inside the dry stock reported in the diagram of Figure 15 is already below the threshold value of 10 GΩ. Afterwards, see Figure 15, resistance increases during the first 72 hours, decreases slightly between 72 and 120 hours, increases slightly again up to 360 hours, and then decreases continuously for the rest of the experiment until it reaches the saturation level of approximately 10 MΩ. During the entire test, resistance remains below the 10 GΩ threshold, which is the criterion for heating elements to be sealed.

Samples placed in the climate chamber for 72 hours at 25 °C in 90 % relative humidity showed a lower initial resistance when compared to the samples stored for 24 hours, see Table 4 and Figure 15. The evolution of resistance in this case shows a slightly different pattern while always remaining at a level below that in the samples stored in the climate chamber with the same temperature-humidity conditions for 24 hours only. Electrical resistance in this case increases during the first 72 hours but then decreases continuously for the rest of the experiment. After 720 hours, the resistance remains at the low saturation level just below 10 MΩ. During the entire measurement the resistance stays below the 10 GΩ limit.

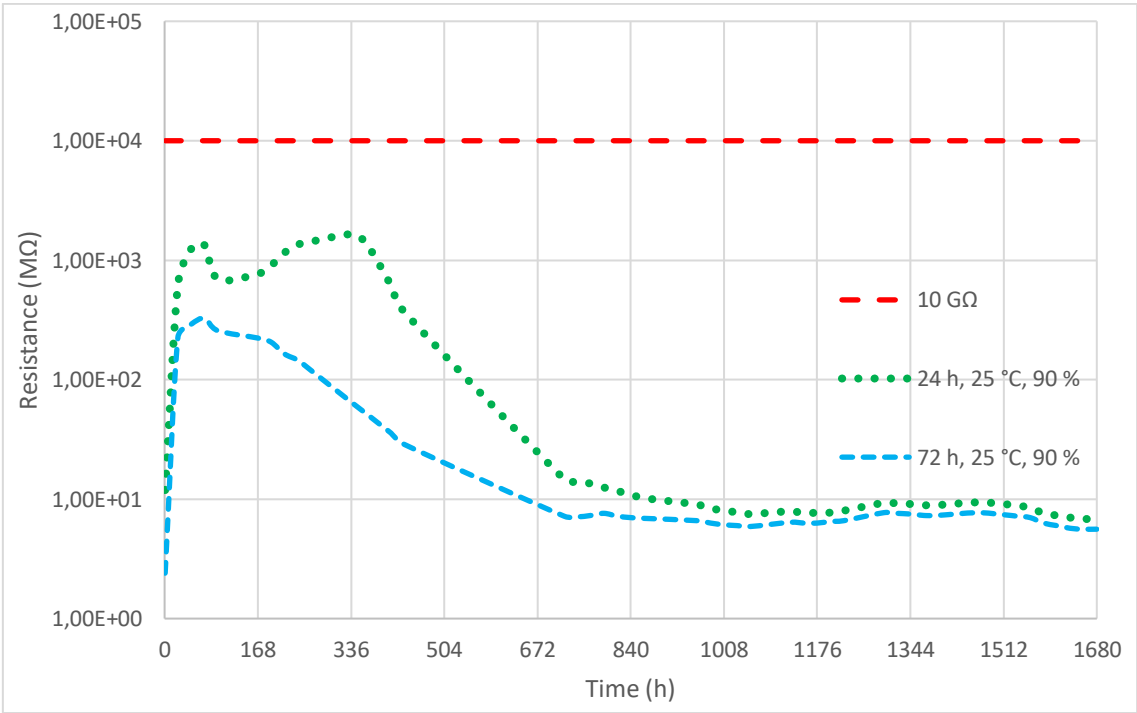


Figure 15: Evolution of electrical resistance of insulation material in annealed tubular heating element samples during exposure to moist air in climate chamber (25 °C, 90 % humidity).

The effect of exposure to hot air with a relatively high level of moisture was studied in the annealed tubular heating element samples by placing them inside the climate chamber at 70 °C in 90 % relative humidity (297 g H<sub>2</sub>O /m<sup>3</sup> air). In this case, electrical resistance degrades to a level well below 10 GΩ upon storing inside the climate chamber and remains at a very low level below 10 MΩ during the entire measurement inside the dry stock. There is no major change in appearance when comparing the resistance for samples stored for 24 or 72 hours. However, the 72-hour stored samples demonstrate lower resistance levels throughout the entire measurement. The samples stored for 72 hours at 70 °C in 90 % relative humidity (297 g H<sub>2</sub>O /m<sup>3</sup> air) demonstrate the lowest initial resistance among all the sample conditions explored in this investigation, see Table 4.

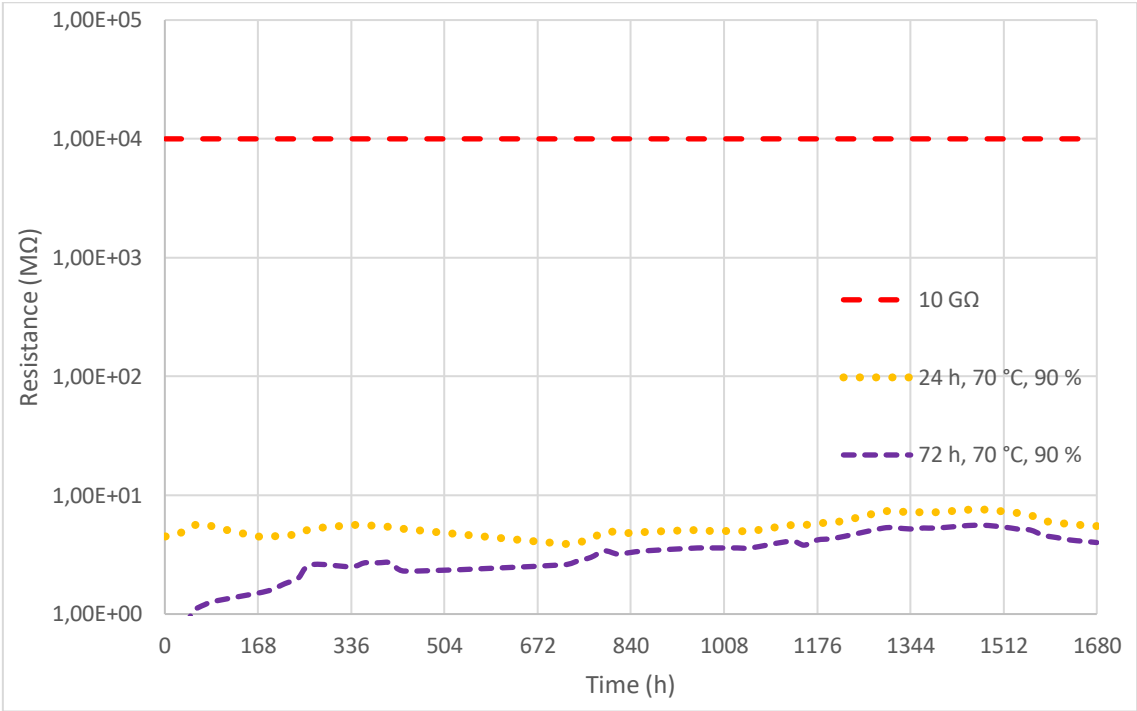


Figure 16: Evolution of electrical resistance of insulation material in annealed tubular heating element samples during exposure to hot and moist air in climate chamber (70 °C, 90 % humidity).

### 3.2 Hydration of MgO insulation from the moisture in ambient air

It is intuitive to expect that the hydration of MgO begins from the edges of tubular heating elements where water contained as moisture in the ambient air penetrates them. The following experiments present the results of investigating such an assumption and the results are summarized in Table 10.

*Table 10: Effect of exposure conditions on the length of insulation material necessary to remove from the ends of a tubular heating element for restoring the level of electrical resistance to an acceptable level above the threshold value of 10 GΩ.*

<b>Time stored (h)</b>	<b>Temperature (°C)</b>	<b>Relative humidity (%)</b>	<b>Resistance after exposure (MΩ)</b>	<b>MgO removed (cm)</b>	<b>Resistance after MgO removal (MΩ)</b>
<b>1</b>	<b>2</b>	<b>3</b>	<b>4</b>	<b>5</b>	<b>6</b>
24	25	30	7400	2	≥11000
48	25	30	3100	4	
72	25	30	1434	6	
24	25	90	1,30	8	
48	25	90	0,7	12	
72	25	90	0,4	14	

The comparison of columns 1-3 and 4 in Table 10 reveals that electrical resistance decreases significantly with the increase of exposure time as well as the level of air humidity. It is also seen that the level of humidity (the concentration of water in ambient air) has a larger effect on resistance than the time of exposure. The comparison of the aforementioned columns with column 5 also suggests that the decrease of electrical resistivity level correlates well with the length of MgO insulation necessary to remove from the edges of tubular heating elements to restore acceptable electrical insulation. These results confirm the expectations as well as suggest that the evolution of MgO hydration depends on both air humidity and exposure time as well as the distance from the edges of tubular heating element.

### 3.3 Effect of sealing material and exposure time

The measurement of electric resistance in tubular heating element samples sealed with PU showed no appreciable dependence on exposure conditions and time. If such samples are sealed immediately after annealing, electrical resistance does not change appreciably during following exposure testing inside the climate chamber in different environments up to at least 72 hours, see Table 11. Resistance remains high during all measurements, see Table 11. The results from the investigation of short-circuit in high voltage testing (1600 V) for PU-sealed samples can also be seen in Table 11.

*Table 11: Effect of exposure conditions on electrical resistance and resistance to short-circuit in high voltage (1600 V) in a tubular heating element sealed with PU.*

Time stored (h)	Temperature (°C)	Humidity (%)	Resistance after CC (MΩ)	Short circuit
24	25	90	≥11000	No
72				
24	70			
72				

The measurement of electrical resistance in tubular heating element samples sealed with Araldite D (epoxy) also showed no appreciable dependence on exposure conditions and time for up to at least 72 hours when samples are sealed immediately after annealing. Resistance observed after the exposure inside the climate chamber can be seen in Table 12. The electrical resistance also remains high during all the measurements, and from the high voltage testing (1600 V) no short circuit is observed, see Table 12.

*Table 12: Effect of exposure conditions on electrical resistance and resistance to short-circuit in high voltage (1600 V) in a tubular heating element sealed with Araldite D.*

Time stored (h)	Temperature (°C)	Humidity (%)	Resistance after CC (MΩ)	Short circuit
24	25	90	≥11000	No
72				
24	70			
72				

The measurement of electrical resistance in tubular heating element samples sealed with Ecobond (epoxy) again showed no appreciable dependence on exposure conditions and time for up to at least 72 hours when samples are sealed immediately after annealing. Resistance observed after the exposure inside the climate chamber in the samples sealed with Ecobond can be seen in Table 13. The resistance remains high during all measurements, and from the high-voltage testing (1600 V) no short circuit is observed, see Table 13.

*Table 13: Effect of exposure conditions on electrical resistance and resistance to short-circuit in high voltage (1600 V) in a tubular heating element sealed with Ecobond.*

<b>Time stored (h)</b>	<b>Temperature (°C)</b>	<b>Humidity (%)</b>	<b>Resistance after CC (MΩ)</b>	<b>Short circuit</b>
24	25	90	≥11000	No
72				
24	70			
72				

These experiments demonstrate that appropriate sealing of tubular heating elements (with similar dimensions) immediately after annealing allows their preservation in varying environmental conditions without the degradation of performance.



### 3.4 Testing of novel UV-curable resins

The measurement of electrical resistance in tubular heating element samples sealed with the novel UV-curable adhesives are summarized in Table 14 along with corresponding exposure conditions. All samples show resistance above 10 GΩ immediately after sealing and before exposure inside the climate chamber. In the testing for 24 hours in 25 °C at 90 % relative humidity (18 g H<sub>2</sub>O /m<sup>3</sup> air), samples sealed with NOA 81 and NEA 121 resins, show no degradation in electrical resistance, see Table 14. By contrast, the samples sealed with Unocol 818 reveal severe drop in electrical resistance due to insufficient curing, resulting in very poor sealing ability and/or adhesion of this resin even at the least humid conditions. Therefore, this resin was excluded from following tests. The samples sealed with NOA 81 and NEA 121 resins demonstrate high resistance in all exposure conditions, and also show no sign of short-circuit when testing with 1600 V is applied.

Table 14: Effect of exposure conditions on electrical resistance and resistance to short-circuit in high voltage (1600 V) in a tubular heating element sealed with UV-curable adhesives.

Adhesive	Time stored (h)	Temperature (°C)	Humidity (%)	Resistance after CC (MΩ)	Current leakage
Unocol 818	24	25	90	5	Yes
	72			-	-
	24	70			
	72				
NOA 81	24	25	90	≥11000	No
	72				
	24	70			
	72				
NEA 121	24	25	90	≥11000	No
	72				
	24	70			
	72				

## 4. Discussion

### 4.1 Importance of insulation in tubular heating elements

The results of electrical insulation measurements in ambient environment can be seen in Table 9 and the storage conditions can be seen in Table 3. These results demonstrate that both silicone plug and humidity influences the electrical resistance of the heating element samples. The silicone plug has a positive effect on the electrical resistance. This is due to decreased amount of MgO in direct contact with ambient air, which limits the access to moisture necessary for the hydration of MgO. This in combination with lower relative humidity gives the highest resistance amongst different samples before annealing. Since the annealing furnace operates at temperatures above 350 °C, the reversed reaction of Mg(OH)<sub>2</sub> to MgO is enabled [10]. This combined with the removal of water vapor gives the samples a high electric resistance after annealing.

The results of electrical insulation measurements in ambient environment can be seen in Figure 14 and the storage conditions can be seen in Table 3. For further details see Appendix I Changes in electrical insulation during exposure to ambient environment. These results demonstrate that electrical resistance decreases rapidly during the first few hours of storage. During annealing, high temperature removes moisture from the heating element, and air inside the samples expand as a result of the increased temperature. This makes the heating elements very dry but also creates vacuum inside the samples. A reason for the latter is that air is forced out of the heating element during annealing due to the elevated temperatures. Upon leaving the furnace, the vacuum sucks surrounding air into the samples and moisture present in the ambient air penetrates the sample very fast. At this point, water vapor inside the heating element acts as an electric conductive substance between the MgO particles [5, 9], which is what causes the rapid decrease in electrical resistance. This further explains the gigantic decrease in resistance when the heating element samples leave the annealing furnace. When studying electrical insulation during the first two hours after annealing, it becomes evident how fast electrical resistance decreases, see Figure 17.

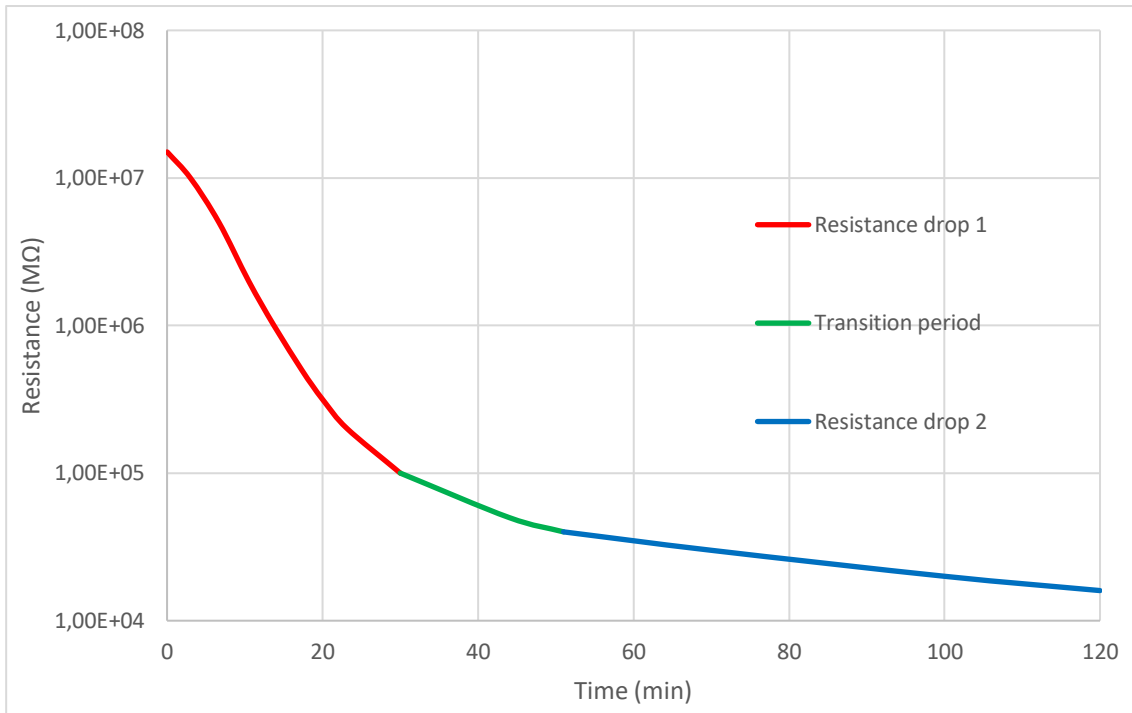


Figure 17: Evolution of electrical resistance of insulation material in annealed tubular heating element samples during the first 2 hours of exposure to moist air in ambient environment.

The decrease in electrical resistance can be split into separate regions. The first (fastest) drop occurs during the initial some 30 minutes where electrical resistance decreases exponentially from  $10^7$  MΩ to  $10^5$  MΩ. This is a gigantic drop in electrical resistance, showing the effect of water vapor acting as an electric conductive substance inside the heating element samples when moisture penetrated [5, 8]. The vacuum created during annealing sucks the ambient air into the heating element, carrying moisture into the heating element at a faster rate. This continues for as long as the pressure inside the heating element is lower compared to that of the ambient environment (atmospheric pressure), which is believed to be the time of the first resistance drop, see Figure 17. This would explain why the penetration of moisture is faster immediately after annealing, resulting in a major decrease in electrical insulation.

Another factor that might influence the process is the difference in humidity levels inside and outside of the heating element sample. Annealing makes the samples completely dry, and thus environment inside samples immediately upon leaving the furnace is less humid in comparison to the ambient air. Since environments move towards equilibrium, moisture enters the heating element until humidity levels outside and inside of the sample become equalized. This work in both ways and if a lot of moisture has penetrated the sample there is a possibility that equilibrium hinders water vapor from entering the sample. This most likely has a minor effect compared to the vacuum effect and the MgO being hygroscopic. However, it is still worth mentioning and could be the reason for the transition period where the rate of decreasing resistance becomes lower compared to the initial 30 minutes.

The following resistance drop decreases exponentially from  $10^5$  MΩ to the threshold of  $10^4$  MΩ (10 GΩ) during the following four hours. The decrease in electrical resistance then continues in a similar rate for 24 hours until it eventually stops. This decrease in resistance can be explained by moisture penetrating the sample as a consequence of the MgO being hygroscopic [7, 10].

The rate of the second resistance drop is lower compared to the first suggesting that the pressure has stabilized inside the heating element at this stage. If pressure levels are the same inside and outside the sample, there is no suction of ambient air into the sample. However, since the heating element has no sealing, and the hygroscopic MgO attracts moisture from the ambient air, there is a continuous flow of water vapor into the heating element.

After 24 hours, the trend changes and electrical resistance is increasing during the following 72 hours, see Figure 18. This increase in resistance can be associated with the decrease of moisture level inside the heating element, most likely due to the absorption of water molecules by MgO when converting into Mg(OH)<sub>2</sub>. The formation of Mg(OH)<sub>2</sub> initiates at temperatures below 350°C as soon as water vapor enters the heating element [5, 10]. However, it is difficult to detect the presence of Mg(OH)<sub>2</sub> until the quantity becomes sufficient enough to affect electrical resistance of the heating elements samples. Initially, the drop of electrical resistivity is controlled by the rapidly increasing amount of most electric conductive component in the sample, which is the moist air (water vapor). However, as the intake of moist air saturates at approximately 24 h upon reaching the pressure balance, and electrical resistivity reaches its local minimum value because of that, another competing process becomes significant. Namely, hygroscopic MgO absorbs water molecules from the moist air inside the heating element thus decreasing the amount of most electric conductive component in the system. The absorption of water molecules when MgO converts into Mg(OH)<sub>2</sub> has a drying effect on the heating element thus altering the trend and increasing electrical resistance [7-9].

After 96 hours the evolution of electrical resistance changes the trend again. It alters to decreasing again, now for the rest of the experiment, see Figure 18. This decrease in resistance can be explained by the increasing hydration of MgO, i.e. the formation of Mg(OH)<sub>2</sub>, along with the following hydration of the latter. Mg(OH)<sub>2</sub> has lower electrical resistivity than MgO (see Table 1) and is also hygroscopic. Therefore, it has the ability to bind water by absorbing it from the ambient air (hydration). Upon hydration, Mg(OH)<sub>2</sub> has the ability to carry ion exchange along particle surfaces and in grain boundaries, which further increases its electrical conductivity [5, 9, 10]. As time progresses more and more Mg(OH)<sub>2</sub> forms and then hydrates, which leads to continuous decline in resistivity. When exists in a sufficient amount, the hydration of Mg(OH)<sub>2</sub> becomes the most critical factor determining the electric insulation of heating element. In this case the hydration of Mg(OH)<sub>2</sub> seems to become dominant after 96 hours.

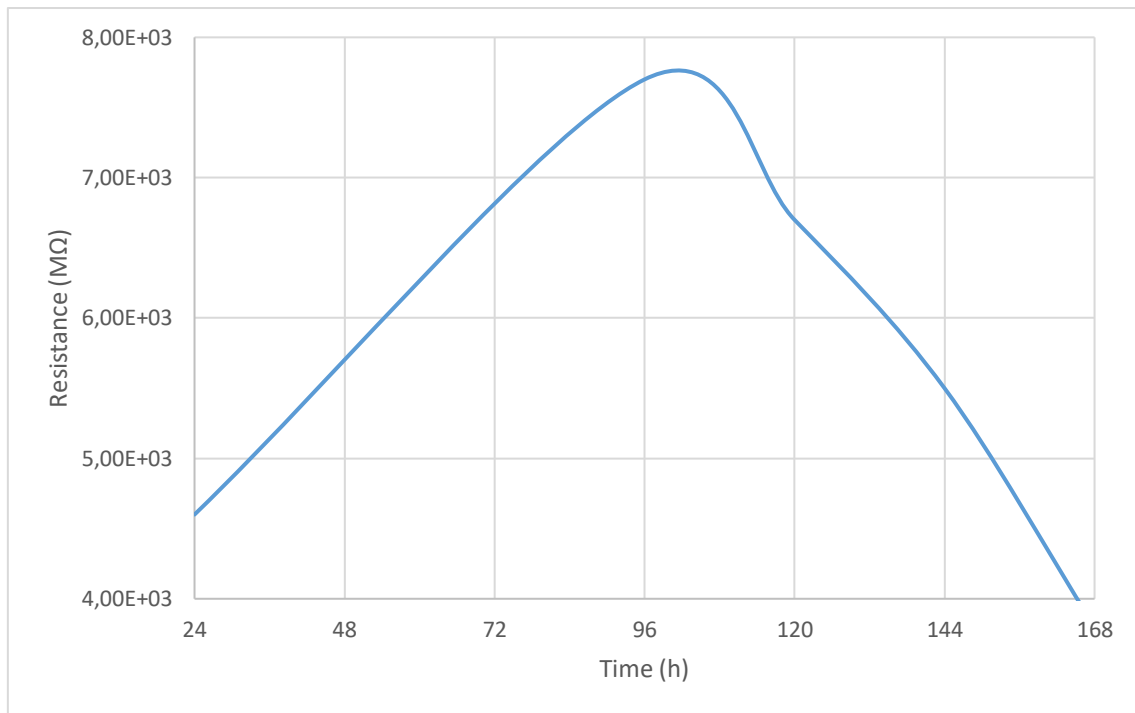


Figure 18: Evolution of electrical resistance of insulation material in annealed tubular heating element samples after pressure stabilization during exposure to moist air in dry stock (25 °C, 30 % humidity).

Since both MgO and Mg(OH)<sub>2</sub> are hygroscopic and therefore have the ability to absorb water from ambient air, electrical resistance can be influenced by the hydration of both MgO and Mg(OH)<sub>2</sub>. This explains why electric resistance both increase and decrease during the measurements, see Figure 14. The alteration of trends in the evolution of electrical resistance over time is due to the following factors:

1. Increase of air humidity (penetration of H<sub>2</sub>O into MgO powder) → resistance decreases.
2. Absorption of H<sub>2</sub>O by MgO (dehydration of air) → resistance increases.
3. Hydration of MgO (formation of Mg(OH)<sub>2</sub>) → resistance decreases.
4. Hydration of Mg(OH)<sub>2</sub> (electrical conductivity of Mg(OH)<sub>2</sub> increases) → resistance decreases.

These four factors coexist during the experiment. The first one dominates in the early stages of measurements when pressure equalizes, and moist air penetrates heating elements (resistance decreases). After 24 h, this process diminishes, and the second factor becomes dominant thus leading to the increase in electrical resistance of the heating elements. At this stage the quantity of Mg(OH)<sub>2</sub> in the system is too small to affect electrical resistance readings. However, as time progresses hydration of MgO leads to the formation of large Mg(OH)<sub>2</sub> quantities, the third factor. After 96 h, this results in the prevalence of the third factor, and further enhancement of the effect by the fourth one. This explains the new decline in resistance over time. Since the elements have no sealing in this experiment, the access to water vapor for the electric insulating material is virtually unlimited. This results into the flow of water vapor into the sample since both MgO and Mg(OH)<sub>2</sub> are hygroscopic [5, 10]. As time progresses more and more MgO converts into Mg(OH)<sub>2</sub> which eventually exist in a sufficient amount inside the sample. At some point, the formation and hydration of Mg(OH)<sub>2</sub> leads to creating an electrical conductive path between the resistance wire and the mantle enabling current leakage, which results in bad electrical insulation. When this happens, the resistance of heating element is no longer affected significantly by the conversion of MgO to Mg(OH)<sub>2</sub> since current leakage

is already enabled. This also suggests that although the quantity of  $\text{Mg(OH)}_2$  formed is important, the distribution of the  $\text{Mg(OH)}_2$  particles in the heating element is important even more. At some stage, the formation of more electrical conductive and hygroscopic  $\text{Mg(OH)}_2$  particles leads to a percolation effect (the theory of how a liquid or gas flows through a porous media) [42]. It is difficult to predict the exact positioning of the  $\text{Mg(OH)}_2$ . But the probability of an electric conductive cluster formation increases the increase of  $\text{Mg(OH)}_2$  quantity. Furthermore, since air enters heating element tubes from their endings, it is expected that the presence of  $\text{Mg(OH)}_2$  is larger in their vicinity. This proves the necessity of a sealing since an electrical conductive path of hydrated  $\text{Mg(OH)}_2$  leads to current leakage, resulting in bad insulation of the heating element.

In order to avoid low electrical resistance, drying operations are necessary in the production cycle. Furnaces for that typically operate at different temperatures up to 200 °C but the temperatures above 350 °C are required in order to reverse the conversion of  $\text{Mg(OH)}_2$  to  $\text{MgO}$  [10]. Temperatures up to 200 °C are still sufficient for removing moisture from the elements. This can be explained by the removal of free water from  $\text{Mg(OH)}_2$ . During hydration,  $\text{Mg(OH)}_2$  binds water along particle surfaces and in grain boundaries. Such water molecules are not bound as strongly as when  $\text{MgO}$  absorbs water when forming  $\text{Mg(OH)}_2$  [5, 7, 10]. This makes it possible to remove water molecules from the  $\text{Mg(OH)}_2$  by heating the heating elements to elevated temperature. This process may not convert the  $\text{Mg(OH)}_2$  back to  $\text{MgO}$  but by removing water molecules from the particle surfaces the electric conductivity of  $\text{Mg(OH)}_2$  decreases resulting in an increased electrical resistance [8, 9]. This explains why heating below 350 °C can still be sufficient to reduce electric conductivity of heating elements and often achieving sufficiently good electrical insulation.

Nevertheless, if hydration of  $Mg(OH)_2$  occurs to a large extent, it becomes difficult to remove water to an extent sufficient for a good electric insulation. At this point, it is difficult to dehydrate  $Mg(OH)_2$ , and either longer furnace time or higher temperatures are required. If happened, it might be better to use the annealing furnace where the temperature is significantly higher. This does not only dehydrate  $Mg(OH)_2$  but will also reverse the chemical reaction converting  $Mg(OH)_2$  to  $MgO$  [10]. However, the converting of  $Mg(OH)_2$  to  $MgO$  is an aggressive reaction, and the removal of water can therefore damage the structure of the insulating material. It is expected that the insulating properties of a once “moisture damaged” heating element is therefore not as good compared to a heating element where moisture penetration was avoided, knowledge provided by Backer AB. This further proves the necessity of improving the sealing process to avoid such in the future.

A comparison of the results from electric insulation in ambient environment can be seen in Figure 19. All samples have low resistance upon leaving storage inside the climate chamber and placed inside the dry stock. The presence of water vapor in large quantity inside the climate chamber enables moisture penetration of the samples resulting in low electrical resistance due to the high presence of water vapor (see Table 4). This also enables larger formation of  $Mg(OH)_2$ , resulting in increased hydration of  $Mg(OH)_2$  and decreasing electrical insulation of the samples.

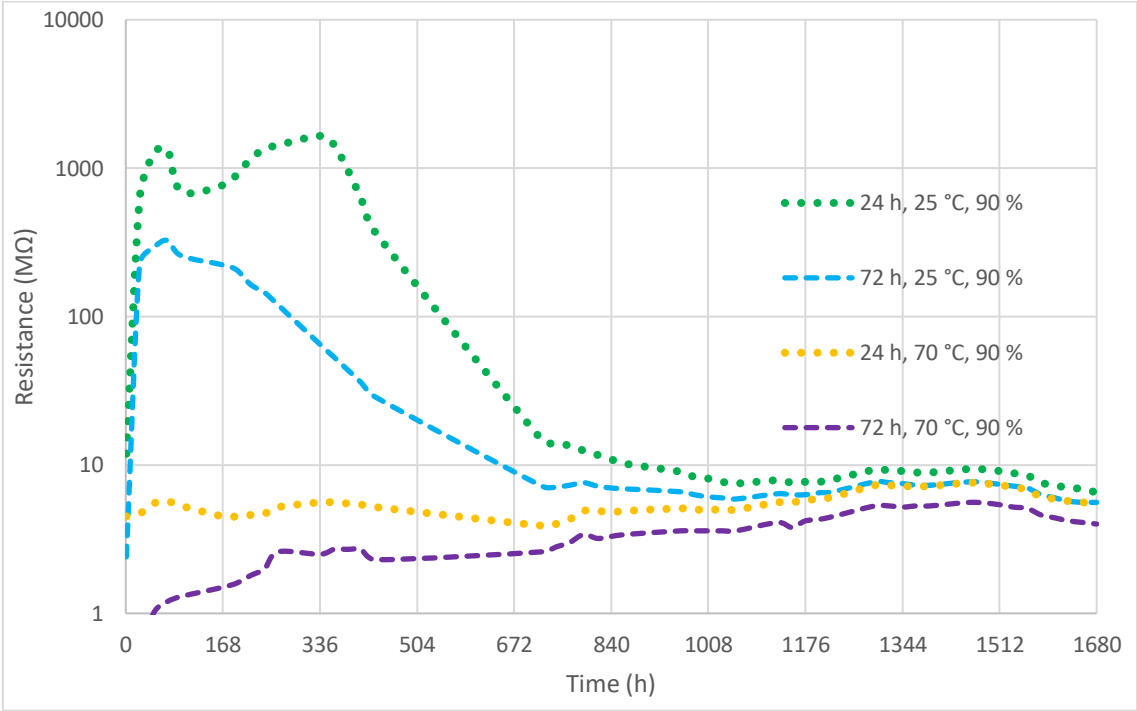


Figure 19: Comparison of evolution of electrical resistance of insulation material in annealed tubular heating element samples during exposure to moist air in climate chamber (25 °C and 70 °C, 90 % humidity).

When examining the behavior of samples stored at 70 °C (second and third curve from above) in Figure 19, it becomes evident that the electrical resistance of these samples is much lower compared to the samples stored at 25 °C (first and second curve from above). When comparing the initial values of resistance after storage inside the climate chamber with values after 72 hours inside the dry stock, it becomes clear that there is a major difference in resistance change, see column 2 and 3 in Table 15.

*Table 15: Resistance of insulation material in samples after exposure to ambient air in climate chamber and after 72 hours of storage in dry stock (25 °C, 30 % humidity).*

<b>Curve color in Figure 14</b>	<b>Initial resistance after CC (MΩ)</b>	<b>Resistance after 72 hours in DS (MΩ)</b>
<b>1</b>	<b>2</b>	<b>3</b>
Green	11,9	1333
Blue	2,4	326
Yellow	4,5	5,7
Purple	0,4	1,2

The amount of absorbed water vapor is the largest in samples stored for 72 hours at 70 °C in 90 % relative humidity (297 g H<sub>2</sub>O /m<sup>3</sup> air) resulting in the lowest initial resistance among the samples. Electrical resistance then remains lowest amongst the different samples during the measurement period. Two factors can explain the lowest electrical resistance of this sample. The first one is the exposure to humid ambient air inside the climate chamber. At 70 °C and 90 % relative humidity water content in the ambient air is the highest, see respective labels in Figure 1. When compared to the water content at 25 °C in 90 % humidity (18 g H<sub>2</sub>O /m<sup>3</sup> air), it is obvious that the presence of moisture is larger inside the samples stored at 70 °C and 90 % relative humidity (297 g H<sub>2</sub>O /m<sup>3</sup> air). The larger presence of moisture then results in a low initial electrical resistance. The second factor is that temperature does not only affect possible amount of water vapor present in the ambient air but also the rate of conversion from MgO to Mg(OH)<sub>2</sub>. At higher temperatures, the rate of conversion increases resulting in a larger presence of Mg(OH)<sub>2</sub> in a shorter period of time [10, 12]. Since more Mg(OH)<sub>2</sub> is present, hydration of such can initiate at an earlier stage of the measurement. As mentioned earlier, if Mg(OH)<sub>2</sub> is forming in a sufficient amount between the resistance wire and the mantle, it creates an electrical conductive path, which enables current leakage. In the case of samples stored for 24 and 72 hours at 70 °C in 90 % relative humidity, it is likely that this has already happened upon leaving the climate chamber. This explains why electrical resistance is very low in the case of both samples during the entire measurement, see Figure 19.

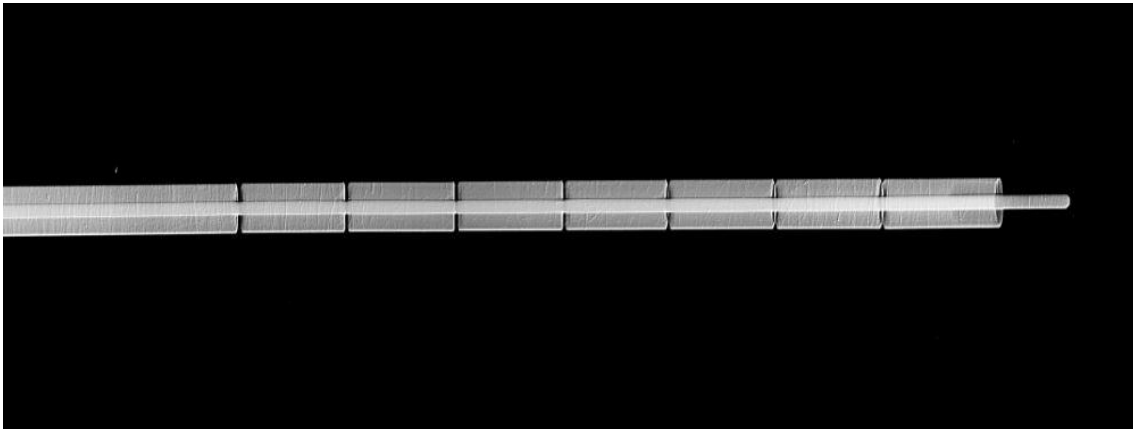


Both samples stored at 25 °C show increasing electrical resistance for approximately 72 hours, see Figure 19. After that, the electrical resistance of the sample stored for 24 hours is higher compared that of the sample stored for 72 hours, under same conditions. This is due to the shorter time of exposure to humid air. Shorter exposure time results in smaller quantity of MgO converting into Mg(OH)<sub>2</sub> and preventing the hydration of the already formed Mg(OH)<sub>2</sub>. When stored for 24 hours at 25 °C in 90 % relative humidity, there is a small decrease in resistance between 72 and 120 hours. This decrease can be explained by the additional penetration of moisture into the sample since both MgO and Mg(OH)<sub>2</sub> are hygroscopic. At this point, the existing amount of Mg(OH)<sub>2</sub> is not yet in a sufficient amount where hydration of such have a major impact on electrical resistance. This can further explain why electrical resistance is increasing up to 360 hours for the sample. At this point the amount of existing Mg(OH)<sub>2</sub> in the sample is sufficient to conduct an electric current when hydrated, and therefore resistance is decreasing for the rest of the measurement. For the sample stored for 72 hours with same conditions, the decrease in electrical resistance initiates after 72 hours of the measurement suggesting that the amount of existing Mg(OH)<sub>2</sub> at this point is sufficient to create an electrical conductive path between the mantle and the resistance wire. This appears logical, as longer exposure to humid air in the climate chamber should result in the formation of larger Mg(OH)<sub>2</sub> quantity. This further results in an earlier decrease of electric resistance as Mg(OH)<sub>2</sub> hydrates.

As already mentioned, temperature plays an important role, as the hydration of MgO converting it to Mg(OH)<sub>2</sub> takes place faster at elevated temperature [12]. The result of faster conversion can be seen when examining different appearances of different samples in Figure 19. Another factor that plays a major role in the kinetics of hydration reaction of MgO to Mg(OH)<sub>2</sub> is the crystallinity of MgO particles [10, 12]. This has not been investigated in this project but previous studies has demonstrated that crystallinity of periclase (MgO phase) does have an effect on the reaction rate when forming Mg(OH)<sub>2</sub> [9, 10, 12]. During experiments in this project the MgO powder is of the same origin for all samples and therefore should not have a different effect on the reaction rate of the MgO. However, in the future investigation of such could be of great interest as it may be helpful to decrease the rate of reaction when MgO is converting into Mg(OH)<sub>2</sub>.

#### 4.2 Effect of hydration of MgO insulation from the moisture in ambient air

When studying the penetration depth of moisture in the samples, the results show that the degree of moisture is decreasing towards the center of the heating elements. This was expected since air entering from each end of the heating element and water molecules are absorbed by the hygroscopic MgO and  $\text{Mg}(\text{OH})_2$  as air flows through the porous solid of MgO. The phenomena where a liquid or gas moves through a porous phase is called percolation. The absorption of water molecules inside the sample is therefore completely stochastic, and it is difficult to determine the distribution of water binding [5, 42]. However, since the first contact between moisture and MgO is at the element ends, the porous network of MgO will act as a filter absorbing water molecules as they come in contact with either MgO or  $\text{Mg}(\text{OH})_2$ . When air is flowing through this network, more and more water molecules become absorbed. This explains why moisture is present in larger quantity close to the endings of the samples but not found to the same extent close to the center. In Appendix II Hydration of MgO insulation from the moisture in ambient air the change in electrical resistance can be examined as MgO is removed from the ends of the samples. In Figure 20, an X-ray image of a heating element where moisture depth analysis has been performed can be seen.



*Figure 20: X-ray image of a heating element with a prolonged bolt and no sealing where MgO has been removed in order to determine depth of moisture penetration.*

Since the samples have no sealing, and both MgO and  $\text{Mg}(\text{OH})_2$  are hygroscopic, moisture continues to flow into the sample. The rate at which MgO converts into  $\text{Mg}(\text{OH})_2$  is affected by different factors such as time of exposure to humid air, humidity levels and temperature [5, 10, 11]. When studying the results in Table 10 and Appendix II Hydration of MgO insulation from the moisture in ambient air it is possible to determine that temperature and humidity have more influence on the depth of moisture penetration compared to the time of exposure. Since temperature affects both the conversion rate of MgO to  $\text{Mg}(\text{OH})_2$  and the possible content of water in ambient air, it is believed to be a good assumption that temperature is the most critical parameter. The depth of moisture penetration at 70 °C exceeded the length of the bolt making it impossible to determine the depth until resistance was above the threshold of 10 G $\Omega$ . However, these results still show the major influence of temperature where increasing the content of water and enabling faster conversion rate allows moisture to penetrate deep into the sample.

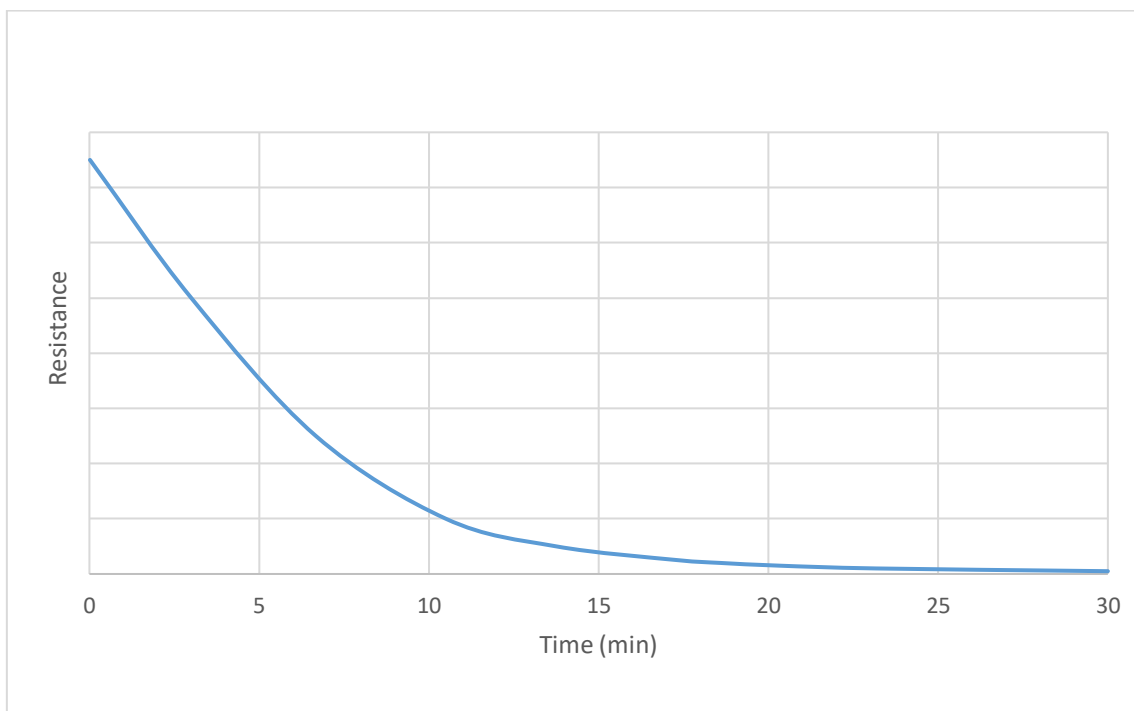
When comparing the effects of exposure time and humidity, it is possible to see that humidity plays a bigger role. One way to see this is to compare sample stored for 72 hours (25 °C, 30 %) and the one stored for 24 hours (25 °C, 90 %). Since temperature is constant, water content is three times higher at 90 % relative humidity compared to 30 %, but the time of exposure is three times longer at 30 % relative humidity. In theory, these two samples should be exposed to the same amount of moisture but when comparing the resistance after exposure to ambient air inside the climate chamber there is an obvious difference in resistance, see column 4 in Table 10. The resistance after 24 hours exposure to 25 °C in 90 % humidity air (1.30 MΩ) is significantly lower compared to the resistance after storage for 72 hours at 25 °C in 30 % humidity air (1434 MΩ). An explanation to this is that moisture penetrates into the sample at a faster rate immediately after the annealing. As mentioned earlier a vacuum is created during annealing, which forces ambient air into the sample at a faster rate once the sample leaves the annealing furnace. At higher relative humidity, air is richer in water molecules, which enables water to travel further into the porous network of MgO before it is consumed. This helps explaining why relative humidity has a larger effect on moisture penetration depth rather than the time of exposure to ambient air inside the climate chamber.

This seems to be a critical point of the process, and water content plays a major role in the amount of moisture that penetrates into the sample while time of exposure to ambient conditions plays a minor role at early stages. This also suggests that if given enough time, the electrical insulation of heating element will decrease towards a similar electrical resistance. This is supported by the trends observed in Figure 19 where resistance eventually becomes similar in all samples.

### 4.3 Effect of sealing material and exposure time

Different samples sealed immediately after annealing do not show any major dependence on exposure conditions and time. No sign of electric leakage or short circuit is observed for samples sealed with PU, Araldite D or Ecobond. However, recent investigations carried out at Backer showed that prematurely sealed heating elements experience current leakage. This seems rather confusing since no such effect has been observed in this study. When comparing the results from these two studies, it became obvious that there was a major difference between the samples of the two studies. In the study carried out in this project, the samples are short (600 mm) and have a relatively large diameter (8.5 mm). In the other study heating elements were much longer and sometimes also possessed a smaller diameter (6.4 mm). This suggests that the dimensions of the element have a major influence on time allowing reliable sealing of heating elements.

At this point there is a strong belief that annealing creates vacuum, which sucks ambient air into the sample. When examining the appearance of curve in Figure 17, it is likely that the lifetime of vacuum for samples with dimensions of 600 x 8.5 mm is close to 30 minutes. Re-plotting this diagram in normal y-axis scale for the first 30 minutes of measurement only, makes obvious that during the lifetime of vacuum, electric resistance drops dramatically, see Figure 21. It is possible to see in Figure 21 the relative difference in electric resistance during the first 30 minutes of the measurement, demonstrating the extent of the first resistance drop from Figure 17.



*Figure 21: Evolution of relative decrease in electrical resistance of insulation material in annealed tubular heating element samples during the initial 30 minutes of exposure to moist air in ambient environment.*

This drop in electrical resistance most likely depends on the dimensions of heating element. Since the MgO network is porous, there are parts of heating element that consist of free space (voids). During annealing, the removal of air from these voids creates a vacuum (low pressure). Upon leaving, the furnace air re-enters the heating elements by attempting to refill the voids, and by doing so stabilizing the pressure inside the heating element. The size or lifetime of vacuum is therefore dependent on the volume of free space inside the heating elements. A heating element with a large volume will therefore create a larger vacuum, during the annealing, due to larger volume of free space inside (more voids in the MgO network). The air enters from each end of the heating element where the diameter acts as the entrance for humid air penetrating the elements.

Results from previous studies carried out by Backer showed that most heating elements that showed signs of current leakage had small diameter relative to the length of the element. This suggests that a combination of large volume and small diameter increases the risk of current leakage if heating elements are sealed prematurely. This makes it possible to explain why heating elements with certain dimensions need time to stabilize their pressure before sealing can be carried out. It can also be of relevance that a small diameter will decrease the distance between the mantle and the resistance wire, thus decreasing the amount of  $\text{Mg}(\text{OH})_2$  necessary to form an electric conductive path between them.

Another theory discussed in this project is that if a heating element with a large free volume is sealed prematurely, it will trap a vacuum inside. When active, the lack of pressure enables ionization of gases, which may lead to an increased electrical conductivity of such heating elements, resulting in higher electric conductivity inside the heating element [43, 44]. In the case of this study, the small size of the heating element samples prevented trapping this since the lifetime of vacuum is short (30 minutes), and pressure stabilizes by the time when heating elements were sealed. This is just a speculation at this point, which requires further elaboration for proving its validity. Yet, it is still worth mentioning as it may explain the reason of current leakage if heating elements are sealed prematurely.

#### 4.4 Improvement and challenges after testing of novel UV-curable resins

The procured adhesives NOA 81 and NEA 121 are both easy to apply, and curing is done in a matter of seconds when exposed to the UV light. There were difficulties with applying Unocol 818, as it did not flow out evenly, which made it hard to cover the surface of MgO powder. One reason could be the higher viscosity of Unocol 818, which is almost twice as thick as NOA 81 and NEA 121 resins (see Table 8). The thickness of Unocol 818 also made curing less efficient, as the penetration depth of UV light was not sufficient to cure sealing mass entirely. The resin cured at the surface but remained in a liquid state underneath. In an attempt to decrease viscosity, Unocol 818 was heated but the problems persisted even after heating the resin. This shows that viscosity is important when making a resin suitable for sealing heating elements. When compared to other resins used by Backer, the thickness of material before curing often becomes an issue as it prevents efficient applying of the sealing material. Results from the investigation of sealing ability show that NOA 81 and NEA 121 are both successful in preventing moisture from penetrating the sample. This is not the case for Unocol 818, as resistance drops after storage in humid conditions, see Table 14. This demonstrates the importance of complete curing and the ability to completely cover the surface of MgO powder. As already mentioned, viscosity is one important parameter to achieve that. A low viscosity makes it possible for the material to cover the surface more evenly and also to fill out surface defects on the mantle and MgO. When exposing material to UV-radiation, low viscosity makes it easier for the light to penetrate through the material and thus to achieve curing throughout the entire sealing. Hence, viscosity is one important parameter when choosing appropriate sealing material.

Further testing of NOA 81 and NEA 121 revealed their capability to prevent moisture from penetrating the samples in temperatures up to 70 °C with a relative humidity of 90 % (297 g H<sub>2</sub>O /m<sup>3</sup> air). At this point, the amount of water vapor in the ambient air is very high, which suggests that these two resins are very good in preventing moisture penetration. However, both adhesives are limited by their temperature range as they have an upper temperature limit of 110 °C (see Table 8). When testing their temperature limit at 120 °C, neither adhesive remained in their cured state. This creates a problem for their utilization since many heating element applications has operating temperatures above 120 °C. Finding UV-curable adhesives that can withstand higher temperatures is therefore of interest. The results from UV-curing testing demonstrate that this is an effective way for reducing curing time and the sealing ability of UV-curable adhesives is very good even when applied in small amounts.

There is one aspect, which can become a problem if proceeding with UV-curing in the sealing process, which is the necessity of using an insulation pearl. The main function of the insulation pearl is to increase the electrical creeping distance and preventing current leakage. The distance an electric current needs to travel between the bolt and the mantle can thus be increased by the addition of the insulating pearl. Such a pearl is attached during curing, which becomes a problem as it limits the entry of light required for UV-curing. This problem was given a consideration in this project, and a suggested way for solving this problem is to change the design of such a pearl. If using a hollow or see-through pearl, light can still reach the sealing material, which is a necessity during curing. Other ways to solve this problem is to combine the use of a UV-curable sealing with another type of sealing material when attaching the pearl. The UV-cured part acts as the main protection towards moisture while the remainder is mainly used for attaching the insulation pearl. One such material could be RTV since it cures at low temperature while can withstand high temperatures. In Figure 22, the tentative design of such a sealing can be seen.

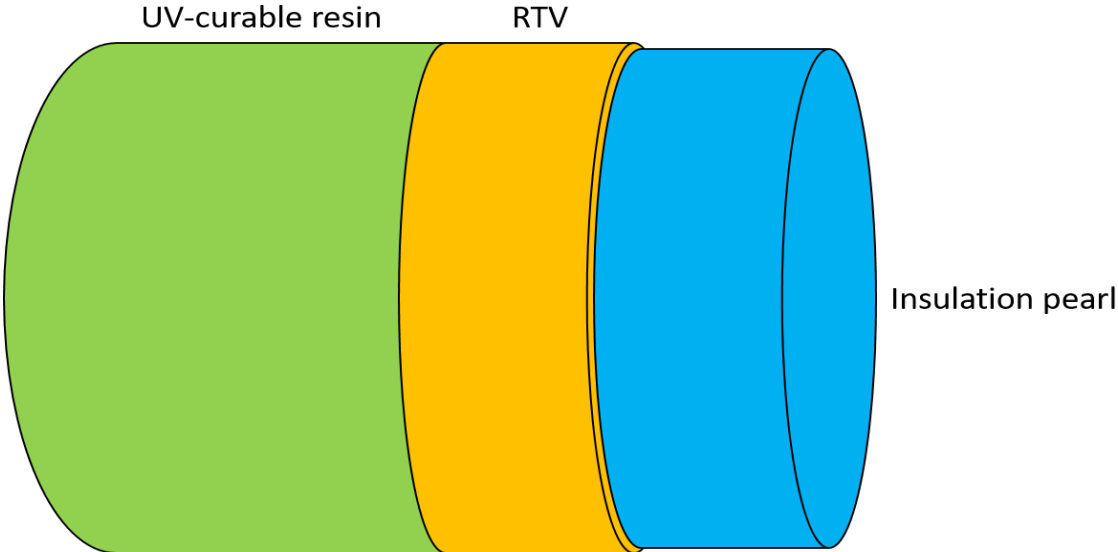


Figure 22: Design of a sealing utilizing both a UV-cured resin and RTV for attaching the insulation pearl.

#### 4.5 Additional comments

During storage inside the dry stock and the climate chamber ambient conditions are assumed to be stable. In reality, this was not the case since the removal of samples from climate chamber or entering and leaving of the dry stock affects the conditions inside. This has a bigger impact on the condition of climate chamber since the space is significantly smaller and the contrast between conditions inside and outside of the climate chamber are larger. When opening the climate chamber door, humidity drops dramatically but this is assumed to be negligible as the climate chamber restores its conditions quickly. The door is only opened when placing or removing samples from the climate chamber making the conditions to remain stable throughout the time of exposure.

In the dry stock, the door opens more frequently as there is more activity compared to using of the climate chamber. However, the space inside the dry stock is much larger and therefore is believed to be less affected by the opening and closing of the entrance door. Also, the samples are stored far away from the door in the dry stock and close to environment-controlling valve, which makes the opening and closing of the door to have little effect on ambient conditions in the samples vicinity.

Another hard to control conditions is when moving samples between annealing furnace, climate chamber and dry stock. As mentioned in the report, vacuum created during annealing has a major effect on the resistance drop, and most likely conditions after the annealing furnace play a big role. To minimize this effect, samples were moved as soon as possible after annealing but some exposure to ambient conditions was difficult to avoid. In order to avoid such, environmental control after the annealing furnace is therefore a necessity. In addition, since measurement of resistance above 11 G $\Omega$  took place in ambient environment in the factory electrical resistance may have been affected. The effect of such exposure is difficult to evaluate but considering the design of the entire process, heating elements experience ambient conditions inside the factory during transport between different stages of the fabrication process. Therefore, this effect is likely experienced in any circumstances.

Finally, the formation of Mg(OH)<sub>2</sub> from MgO is an exothermal reaction, which means that heat is released when reaction occur. This might have a small effect on the formation of Mg(OH)<sub>2</sub> locally inside the heating element samples. Since previous studies show that the conversion rate of MgO to Mg(OH)<sub>2</sub> increases at higher temperatures [10, 12], it is possible that heat released during the formation of Mg(OH)<sub>2</sub> can trigger this reaction further. However, this is believed to have a minor effect on the results of this study, and no major heat release was observed during measurements.



## 5. Conclusions

In this project, the sealing of tubular heating elements has been investigated in order to create a better platform for the optimization of their fabrication process. One of the main interests was to investigate how MgO reacts in the presence of moist air. The sensitivity of MgO to moist air is one of the main reasons for sealing heating elements. Samples for the investigation consisted of short elements (relatively low length/diameter ratio) produced according to company standards. Testing involved the measurements of electric insulation, investigation of hydration of MgO and testing of novel UV-curable resins.

The investigation revealed a major drop in resistance immediately after annealing of heating elements, which is caused by the penetration of moist air into the samples. The penetration of moist air is dictated by the creation of a vacuum in the tubular heating elements during annealing. The exposure of annealed samples to humid environments demonstrated that both air humidity and temperature have major effect on electrical insulation in heating elements. After annealing, the electrical conductivity of MgO determines the electric resistance of the heating element. When moisture penetrates the heating element, the electrical conductivities of MgO and water determines the electric resistance of the heating elements. When MgO is hydrated the electrical conductivity of  $\text{Mg(OH)}_2$  determines the electric resistance of the heating elements and further hydration of such results in the electric insulation of heating elements being determined by the electric conductivities of water and hydrated  $\text{Mg(OH)}_2$ .

The study of penetration depth showed that moisture can penetrate deep into the tubular elements but the concentration of moisture is highest in the vicinity of the heating element ends.

UV-curable adhesives are found to be potent sealing materials that can be used to decrease the time of sealing process in spite of evident challenges in the usable temperature range and the efficiency of curing.

## 6. Future work

There are many ways to improve the sealing of tubular heating elements and further investigation of such is needed.

The main focus in this project was to study the behavior of MgO insulation when it is exposed to humid conditions. The study also allows to make projections for future work. The creation of vacuum during annealing is one aspect requiring further investigation of varying dimensions of heating elements on electric insulation. Mitigating its effect is a most likely key in optimizing the sealing process since it enables a possibility to exclude the need for drying operations, making the production more time and energy efficient.

Further investigation of potential new sealing materials is another area where more work is necessary. Finding UV-curable resins that can withstand high temperatures and are compatible with insulating pearl along with studying of viscosity properties and others are of major interest.

A final interesting aspect is to investigate the point of failure for different sealing materials. Many of the materials used today are known to work under different circumstances but little is known about their limitations. Understanding of such should provide further knowledge regarding properties and handling of different sealing materials, which is necessary in optimizing the processes of sealing and achieving good electrical insulation.

## References

1. NIBE. *NIBE Group – a global organisation with companies and a presence worldwide*. 2019; Available from: <https://www.nibe.com/nibe-group.html>.
2. Backer. *BackerGroup*. 2019; Available from: <https://www.backergroup.com/sv-se/om-oss>.
3. ElectronicsTutorials. *Resistivity*. 2019; Available from: <https://www.electronics-tutorials.ws/resistor/resistivity.html>.
4. Osborne, A. *What is the difference between resistivity & conductivity?* 2018-03-13; Available from: <https://sciencing.com/difference-between-resistivity-conductivity-8221785.html>.
5. Karato, S.-i. and D. Wang, *Electrical conductivity of minerals and rocks*. Physics and Chemistry of the Deep Earth, 2013: p. 145-182.
6. Hansen, K.K., et al. *Magnesium-oxide boards cause moisture damage inside facades in new Danish buildings*. in *International RILEM Conference on Materials, Systems and Structures in Civil Engineering*. 2016. Rilem publications.
7. Gasc, J., et al., *Electrical conductivity of polycrystalline Mg (OH) 2 at 2 GPa: effect of grain boundary hydration–dehydration*. Physics and Chemistry of Minerals, 2011. **38**(7): p. 543-556.
8. Fuji-Ta, K., et al., *Electrical conductivity measurements of brucite under crustal pressure and temperature conditions*. Earth, planets and space, 2007. **59**(6): p. 645-648.
9. Wu, X., et al., *Electrical conductivity measurements of periclase under high pressure and high temperature*. Physica B: Condensed Matter, 2010. **405**(1): p. 53-56.
10. Kuenzel, C., et al., *The mechanism of hydration of MgO-hydromagnesite blends*. Cement and Concrete Research, 2018. **103**: p. 123-129.
11. Santos Jr, T., et al., *Mg (OH) 2 Nucleation and Growth Parameters Applicable for the Development of MgO-Based Refractory Castables*. Journal of the American Ceramic Society, 2016. **99**(2): p. 461-469.
12. Gieseke, W. and F. Freund, *The influence of molecular hydrogen on the conductivity of magnesium oxide containing paramagnetic surface defects*. Journal of Physics and Chemistry of Solids, 1977. **38**(2): p. 183-186.
13. *Humidity*. Available from: <https://www.nationalgeographic.org/encyclopedia/humidity/>.
14. Fargas, G., M. Anglada, and A. Mateo, *Effect of the annealing temperature on the mechanical properties, formability and corrosion resistance of hot-rolled duplex stainless steel*. Journal of materials processing technology, 2009. **209**(4): p. 1770-1782.

15. People, E.T.S. *High-Performance Resin Chemistries for Electronics Protection*. 2018; Available from: <https://www.electrolube.com/technical-articles/high-performance-potting-compound-chemistries-for-electronics-protection/>.
16. People, E.T.S. *Conformal Coatings Vs Potting Compounds Which is better to protect my PCB, a coating or a resin?* 2018; Available from: <https://www.electrolube.com/technical-articles/conformal-coatings-vs-potting-compounds-which-is-better-to-protect-my-pcb-a-coating-or-a-resin/>.
17. Masson, F., et al., *UV-Radiation curing of waterbased urethane–acrylate coatings*. *Progress in Organic Coatings*, 2000. **39**(2-4): p. 115-126.
18. Saenz-Dominguez, I., et al., *Effect of ultraviolet curing kinetics on the mechanical properties of out of die pultruded vinyl ester composites*. *Composites Part A: Applied Science and Manufacturing*, 2018. **109**: p. 280-289.
19. Zhou, J., et al., *Fast curing of thick components of epoxy via modified UV-triggered frontal polymerization propagating horizontally*. *Materials Letters*, 2016. **176**: p. 228-231.
20. Abdoli, H., P. Alizadeh, and K. Agersted, *Fabrication and sealing performance of rare-earth containing glass–ceramic seals for intermediate temperature solid oxide fuel cell applications*. *Ceramics International*, 2014. **40**(5): p. 7545-7554.
21. Donald, I., et al., *Recent developments in the preparation, characterization and applications of glass-and glass–ceramic-to-metal seals and coatings*. *Journal of Materials Science*, 2011. **46**(7): p. 1975-2000.
22. people, E.T.s. *Thermal Management Solutions For Long Term Protection Of Military And Aerospace Electronics*. 2018; Available from: <https://www.electrolube.com/technical-articles/thermal-management-solutions-for-long-term-protection-of-military-and-aerospace-electronics/>.
23. Au, K., et al. *High temperature durable hermetic sealing material selection and reliability comparison for IR gas sensor module packaging*. in *Electronics Packaging Technology Conference (EPTC), 2016 IEEE 18th*. 2016. IEEE.
24. Bley, O., J. Siepmann, and R. Bodmeier, *Importance of glassy-to-rubbery state transitions in moisture-protective polymer coatings*. *European Journal of Pharmaceutics and Biopharmaceutics*, 2009. **73**(1): p. 146-153.
25. Jaiswal, P., et al., *Comparative evaluation of sealing ability of light cure glass ionomer cement and light cure composite as coronal sealing material: An in vitro study*. *Journal of the International Clinical Dental Research Organization*, 2017. **9**(1): p. 12.
26. Bruening, M. and D. Dotzauer, *Polymer films: Just spray it*. *Nature materials*, 2009. **8**(6): p. 449.

27. Hu, J., et al., *The freezing process of continuously sprayed water droplets on the superhydrophobic silicone acrylate resin coating surface*. Applied Surface Science, 2014. **317**: p. 534-544.
28. People, E.T.S. *Polyurethane Encapsulants - Enhanced Protection For LEDs in Challenging Environments*. 2018; Available from: <https://www.electrolube.com/technical-articles/polyurethane-encapsulants-enhanced-protection-for-leds-in-challenging-environments/>.
29. Chattopadhyay, D., S.S. Panda, and K. Raju, *Thermal and mechanical properties of epoxy acrylate/methacrylates UV cured coatings*. Progress in Organic Coatings, 2005. **54**(1): p. 10-19.
30. Firdous, H. and M. Bajpai, *UV curable heat resistant epoxy acrylate coatings*. 2010.
31. Yu, Z., et al., *Preparation and properties studies of UV-curable silicone modified epoxy resin composite system*. Journal of applied biomaterials & functional materials, 2018. **16**(1\_suppl): p. 170-176.
32. Dai, S., et al., *Sealing glass-ceramics with near-linear thermal strain, part III: Stress modeling of strain and strain rate matched glass-ceramic to metal seals*. Journal of the American Ceramic Society, 2017. **100**(8): p. 3652-3661.
33. Borhan, A., et al., *Influence of (CoO, CaO, B2O3) additives on thermal and dielectric properties of BaO–Al2O3–SiO2 glass–ceramic sealant for OTM applications*. Ceramics International, 2016. **42**(8): p. 10459-10468.
34. Ammar, A.U., et al., *Electrochemical Study of Polymer and Ceramic-Based Nanocomposite Coatings for Corrosion Protection of Cast Iron Pipeline*. Materials, 2018. **11**(3): p. 332.
35. Poli, T., L. Toniolo, and O. Chiantore, *The protection of different Italian marbles with two partially fluorinated acrylic copolymers*. Applied Physics A, 2004. **79**(2): p. 347-351.
36. Toniolo, L., et al., *Tailoring new fluorinated acrylic copolymers as protective coatings for marble*. Journal of cultural Heritage, 2002. **3**(4): p. 309-316.
37. Liu, F., et al., *Green fabrication of ultraviolet curable epoxy acrylate-silica hybrid coatings*. Progress in Organic Coatings, 2017. **109**: p. 38-44.
38. Wang, T. and H. Zupko, *Long-term mechanical behaviour of optical fibres coated with a UV-curable epoxy acrylate*. Journal of Materials Science, 1978. **13**(10): p. 2241-2248.
39. Fridrichovsky, M., F. Steiner, and M. Hirman. *Comparison of the characteristics of PCB protective coatings*. in *Electronics Technology (ISSE), 2017 40th International Spring Seminar on*. 2017. IEEE.

40. Brachaczek, W., *The modelling technology of protective silicone coatings in terms of selected physical properties: Hydrophobicity, scrub resistance and water vapour diffusion*. *Progress in Organic Coatings*, 2014. **77**(4): p. 859-867.
41. Bley, O., J. Siepmann, and R. Bodmeier, *Characterization of moisture-protective polymer coatings using differential scanning calorimetry and dynamic vapor sorption*. *Journal of pharmaceutical sciences*, 2009. **98**(2): p. 651-664.
42. Stauffer, D. and A. Aharony, *Introduction to percolation theory*. 2014: Taylor & Francis.
43. Holanda, R., *Sensitivity of hot-cathode ionization vacuum gages in several gases*. 1972.
44. *Ionization*. 2016 October 17, 2016 [cited 2019 March 19, 2019]; Available from: <https://www.britannica.com/science/ionization>.

## Appendices

### Appendix I Changes in electrical insulation during exposure to ambient environment

*Table 16: Measured values from evolution of electrical resistance of insulation material in annealed tubular heating element samples during exposure to moist air in dry stock (25 °C, 30 % humidity).*

<b>Time (h)</b>	<b>Sample 1 (MΩ)</b>	<b>Sample 2 (MΩ)</b>
0	3,00E+07	3,00E+07
24	3,20E+03	4,00E+03
48	6,30E+03	6,00E+03
72	5,70E+03	5,60E+03
144	5,80E+03	5,50E+03
168	5,00E+03	4,60E+03
192	4,10E+03	3,70E+03
216	2,80E+03	2,50E+03
240	2,50E+03	2,20E+03
528	1,16E+03	9,20E+02
552	1,67E+03	1,32E+03
576	1,82E+03	1,49E+03
600	1,70E+03	1,44E+03
624	1,32E+03	1,11E+03
648	1,53E+03	1,18E+03
672	1,85E+03	1,38E+03
744	1,72E+03	1,24E+03
768	1,85E+03	1,31E+03
792	1,40E+03	1,08E+03
816	1,26E+03	9,94E+02
840	1,67E+03	1,18E+03
864	1,98E+03	1,45E+03
888	1,80E+03	1,34E+03
912	1,96E+03	1,47E+03
936	2,15E+03	1,70E+03

960	2,14E+03	1,77E+03
1032	1,43E+03	1,53E+03
1056	1,19E+03	1,35E+03
1080	1,07E+03	1,24E+03
1104	8,35E+02	9,90E+02
1128	6,80E+02	8,40E+02
1200	4,32E+02	5,55E+02
1248	2,88E+02	3,73E+02
1272	2,29E+02	2,96E+02
1296	1,84E+02	2,34E+02
1320	1,07E+02	1,32E+02
1344	9,51E+01	1,15E+02
1368	8,36E+01	9,92E+01
1392	6,85E+01	8,02E+01
1416	5,87E+01	6,80E+01
1488	4,18E+01	4,52E+01
1512	3,70E+01	4,05E+01
1536	3,36E+01	3,61E+01
1560	2,93E+01	3,11E+01
1584	2,78E+01	2,97E+01
1656	2,40E+01	2,44E+01
1680	2,25E+01	2,28E+01



Table 17: Measured values from evolution of electrical resistance of insulation material in annealed tubular heating element samples after exposure to moist air inside climate chamber for 24 hours at 25 °C in 90 % humidity.

Time (h)	Sample 1 (MΩ)	Sample 2 (MΩ)
0	11,9	11,3
24	625	820
48	1245	1634
72	1333	1456
144	696	1160
168	768	1110
192	903	965
216	1155	913
240	1350	805
528	1430	667
552	1650	512
576	1442	453
600	973	348
624	613	263
648	369	158,4
672	14,7	11,8
744	14	11,4
768	13,5	11,6
792	12,5	11,4
816	11,7	10,8
840	10,9	10,1
864	10,2	9,6
888	9	8,9
912	8,3	8,4
936	8,1	8,2
960	7,7	7,9
1032	7,5	7,8
1056	7,9	8,4
1080	7,6	8,3

1104	7,7	8,3
1128	7,7	8,5
1200	11,9	11,3
1224	8,0	8,8
1296	9,3	10
1320	9,2	10,8
1344	9,1	10,7
1368	8,9	10,7
1392	8,9	10,8
1464	9,4	11,7
1512	9,1	11,6
1536	8,8	11,2
1560	8,5	11
1584	7,6	10
1608	7,3	9,6
1632	7	9,2
1656	6,9	9,2
1680	6,5	8,9

Table 18: Measured values from evolution of electrical resistance of insulation material in annealed tubular heating element samples after exposure to moist air inside climate chamber for 72 hours at 25 °C in 90 % humidity.

Time (h)	Sample 1 (MΩ)	Sample 2 (MΩ)
0	2,4	3,5
24	226	310
48	291	411
72	326	413
144	257	264
168	223	79,8
192	206	57,7
216	164,3	44,4
240	145,4	38,9
528	119,4	34,5
552	65	25,5
576	53,4	23,4
600	43,7	20,9
624	35,7	18,4
648	28,7	16,1
672	7,2	7
744	7,1	7,1
768	7,3	7,7
792	7,6	8,7
816	7,2	8,2
840	7	8,3
864	6,9	8,2
888	6,6	8,4
912	6,3	8,2
936	6,1	8,2
960	6	8,1
1032	5,9	8,1
1056	6,4	9,3
1080	6,3	9,2

1104	6,3	9,4
1128	6,5	9,8
1200	2,4	3,5
1224	6,6	10,3
1296	7,7	13,3
1320	7,6	13,2
1344	7,5	13,2
1368	7,3	13,2
1392	7,3	13,6
1464	7,7	15,1
1512	7,4	15,2
1536	7,2	15
1560	7	14,6
1584	6,3	13,8
1608	6	13,5
1632	5,7	13,3
1656	5,6	13,2
1680	5,6	13,2

Table 19: Measured values from evolution of electrical resistance of insulation material in annealed tubular heating element samples after exposure to moist air inside climate chamber for 24 hours at 70 °C in 90 % humidity.

Time (h)	Sample 1 (MΩ)	Sample 2 (MΩ)
0	4,5	4,4
24	4,7	4,6
48	5,5	5,4
72	5,7	5,7
144	5,3	5,4
168	4,5	4,6
192	4,5	4,6
216	4,6	4,6
240	4,7	4,8
528	5,2	5,2
552	5,6	5,6
576	5,6	5,7
600	5,5	5,7
624	5,4	5,5
648	5,2	5,2
672	3,9	4,1
744	4	4,3
768	4,3	4,6
792	4,9	5,2
816	4,9	5,2
840	4,8	5,2
864	4,9	5,2
888	5,1	5,5
912	5	5,4
936	5	5,3
960	5	5,3
1032	5	5,4
1056	5,6	6
1080	5,6	6

1104	5,8	6,1
1128	5,9	6,3
1200	4,5	4,4
1224	6,1	6,6
1296	7,3	7,8
1320	7,3	8
1344	7,2	7,9
1368	7,2	8
1392	7,2	8,1
1464	7,6	8,8
1512	7,3	8,6
1536	7,1	8,4
1560	6,8	8,2
1584	6,1	7,5
1608	5,9	7,2
1632	5,7	6,9
1656	5,6	6,9
1680	5,5	6,5

## Appendix II Hydration of MgO insulation from the moisture in ambient air

*Table 20: Measured values from studying of effect of exposure conditions on the length of insulation material necessary to remove from the ends of a tubular heating element for restoring the level of electrical resistance to an acceptable level above the threshold value of 10 GΩ.*

<b>Time stored (h)</b>	<b>Temperature (°C)</b>	<b>Relative humidity (%)</b>	<b>MgO removed (cm)</b>	<b>Resistance (MΩ)</b>
24	25	30	0	7400
			2	≥ 11000
48	25	30	0	3100
			2	6900
			4	≥ 11000
72	25	30	0	1434
			2	1872
			4	3800
			6	≥ 11000

Table 21: Measured values from studying of effect of exposure conditions on the length of insulation material necessary to remove from the ends of a tubular heating element for restoring the level of electrical resistance to an acceptable level above the threshold value of 10 GΩ after storage for 24, 48 and 72 hours (25 °C, 30 % humidity).

Time stored in CC (h)	Temperature in CC (°C)	Relative humidity (%)	MgO removed (cm)	Resistance (MΩ)
24	25	90	0	1,30
			2	1,36
			4	9,30
			6	802
			8	≥ 11000
48	25	90	0	0,7
			2	0,9
			4	1,7
			6	13,4
			8	231
			10	6500
			12	≥ 11000
72	25	90	0	0,4
			2	0,4
			4	0,6
			6	2,2
			8	14,5
			10	160
			12	3400
			14	≥ 11000



Table 22: Measured values from studying of effect of exposure conditions on the length of insulation material necessary to remove from the ends of a tubular heating element for restoring the level of electrical resistance to an acceptable level above the threshold value of 10 GΩ after storage for 72 hours (25 °C, 30 % humidity) and 72 hours (25 °C, 90 % humidity).

Time stored in CC (h)	Temperature (°C)	Relative humidity (%)	Time stored in DS (h)	MgO removed (cm)	Resistance (MΩ)
72	25	90	72	0	11,8
				2	12,6
				4	13
				6	19,3
				8	76
				10	359
				12	2400
				14	≥ 11000

Table 23: Measured values from studying of effect of exposure conditions on the length of insulation material necessary to remove from the ends of a tubular heating element for restoring the level of electrical resistance to an acceptable level above the threshold value of 10 GΩ after storage for 24 hours (70 °C, 90 % humidity).

Time stored in CC (h)	Temperature (°C)	Relative humidity (%)	MgO removed (cm)	Resistance (MΩ)
24	70	90	0	0,5
			2	0,9
			4	3,4
			6	10,3
			8	32,8
			10	107
			12	335
			14	1200
			16	4000

Table 24: Measured values from studying of effect of exposure conditions on the length of insulation material necessary to remove from the ends of a tubular heating element for restoring the level of electrical resistance to an acceptable level above the threshold value of 10 GΩ after storage for 48 hours (70 °C, 90 % humidity).

Time stored in CC (h)	Temperature (°C)	Relative humidity (%)	MgO removed (cm)	Resistance (MΩ)
48	70	90	0	0,7
			2	1,7
			4	4,8
			6	11,2
			8	23,8
			10	45,7
			12	84,6
			14	169,6
			16	308