



## COMPARISON OF PARAMETERS IN THE FORMATION OF CORROSIVE SULPHIDE DEPOSITION ON COPPER CONDUCTORS

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### ABSTRACT

Copper is a catalyst that promotes the formation of corrosive sulphur but it is also corroded by the latter. Corrosive sulfur in oil has been identified as the cause of recent failures in power transformers and shunt reactors. The most common reason of such failures is arcing between adjacent disks or conductors of windings due to the formation of copper sulphide deposition on cellulosic insulating paper. Synergetic effects with temperature, time and oxygen are recognized to play a role in the formation of corrosive sulphur. Which of these factors has the most impact on copper sulphide deposition? To address this concern, a quantitative laboratory technique has been developed. It is shown that by using a series of laboratory experiments in accordance with ASTM D 1275 B, it is possible to investigate this problem and to map the influence of these parameters by manipulating some variables such as oxygen, temperature and time on the copper samples. The obtained results show that temperature is the most influential parameter in the formation of corrosive sulphur. The process is accelerated when both temperature and time act conjointly.

**Keywords:** corrosive sulphur, time, oxygen, temperature, power transformers, experimental design (DOE).

### INTRODUCTION

Power transformers are greatly important to power systems as they are highly loaded and expensive. The consequences of transformer failure can be quite damaging. In the last decade, corrosive sulfur recognized as a major risk to high voltage oil-filled equipment. Problems encountered with copper sulphide in transformer insulation are still issues of concern in transformer industry. According to a recent study, the direct cost of corrosion is estimated to be approximately \$ 276 billion a year in the United States [1]. Figure-1 [2], sketches statistics of the various failures resulting from corrosive sulfur in power transformers. In practice, the corrosion process occurs when copper sulphide created by the chemical reaction between the oil and the copper migrates into the paper. The after effects of corrosive sulfur into transformers can be disastrous, not only adversely affecting the conductor material and other metal surfaces but may also have drastic effects on paper insulation. Sulfur corrosion of copper wires can reduce the dielectric constant of oil-paper insulation [3]. Since copper sulphide is conductive, the electrical breakdown strength of oil-paper insulation with copper sulfide depositions declines greatly, thence leading to internal insulation failure of transformers. Many researchers have focused on solving the problem of corrosive sulfur in insulating oil. The degree of corrosiveness in insulating oil is a function of the type and concentration of the corrosive substance, the surface condition of the copper conductors, the oxygen access, the temperature and time of exposure as well as the type of oil in the transformer [4]. With the increase of

temperature, corrosive sulfur reactions with copper are more active, which means that the temperature promotes the reaction between corrosive sulfur and copper. Decreasing the temperature by 10°C is reported to decrease the rate of copper sulfide deposition by as much as one-half

[5]. Dissolved air in oil is the main source of oxygen; thus, oxygen can increase oil aging and promote the reaction between corrosive sulfur and copper. Higher oxygen content has been documented to aid in slowing the copper sulfide deposition process [6]. The corrosion process is also most likely time dependent and it may take several months or years to manifest [7]. Recent work [3, 8] showed that the amount of Cu<sub>2</sub>S deposition on insulation paper and copper wire surface increase with aging time and temperature. The aim of this paper is to contribute to the resolution of the problem raised by corrosive sulfur.

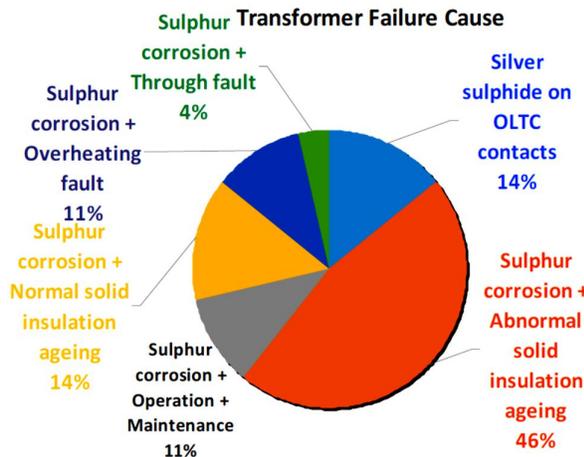


Figure-1. Transformer failures caused by sulphur corrosion.

## BACKGROUND AND REVIEW LITERATURE

### Corrosive sulfur

Corrosion is the spontaneous destruction of metals and alloys caused by chemical, biochemical, and electrochemical interaction between metals and alloys, and the environment. Corrosive environments include moisture, oxygen, inorganic and organics acids, high pressure, temperature, time and chlorides [9]. During corrosion, metals tend to convert to more thermodynamically stable compounds such as oxide, hydroxides, salts, or carbonates:  $Me^0 \rightarrow Me^{n+}$ . The total sulfur content of a mineral oil is formed from a number of sulfurous species with varying reactivities. Crude oil can contain up to 20% sulfur including species such as thiophens, disulfides and polysulfides, thio-ethers, mercaptans and elemental sulfur [7]. Thio-ethers are moderately corrosive compounds which can be found after the oil refining processes with concentration relative to the severity [10]. Disulfides and polysulfides are normally stable compounds but some species such as dibenzyl disulphide (DBDS) can become susceptible to heat breakdown and form mercaptans (R-SH) which are corrosive in transformers. Thiophens are very stable compounds found after all refining processes, which act as natural oxidation inhibitors and are necessary for totally uninhibited oil [10]. Among these compounds, the most studied is the DBDS because it is one of the most active for what concerns corrosion [11]. At low temperatures, DBDS is stable and acts as an oxidation inhibitor but at high temperatures, it becomes corrosive. It has been suggested that DBDS has a triggering temperature, which is known to be below 150°C [12]. A recent paper from CIGRE Working Group A2.40 [2] provides the mechanism of copper sulphide formation illustrated in Figure-2. The formation of  $Cu_xS$  initially begins with the dissolution of copper in oil to form soluble intermediates. According to Figure-2, the following reaction sequences lead to the formation of copper sulphide:

- The occurrence of free and highly reactive copper species at the metal surface, characterized by very limited solubility in oil, possibly due to local thermal excitation or electrical stresses.
- The formation of dissolved copper intermediates, facilitated by the presence of oxygen and some antioxidants, due to the complexation activity of organic components of the oils (e.g. hydroperoxides).
- The displacement/diffusion of the intermediates into the oil and paper insulation.
- The reaction of dissolved copper intermediates with reactive sulfur species to form  $Cu_xS$  as a corrosion by-product on paper.

This complex deposition suggests that several parameters influence the chemical reactions responsible for the copper surface corrosion and subsequent formation of copper sulphide and other deposits on transformer insulation [13]. There are several methods to solve the problem of corrosive sulfur in power transformers. The most commonly used and highly accepted ones are those proposed by ASTM D1275B (copper corrosion) [14] and IEC 62697: 2012 (DBDS quantification) [15], DIN 51353 (silver plate corrosion) and IEC 62535 (corrosion of paper wrapped conductors). Actually, the passivators are more used for solving this problem but do not stop the corrosive effects. The passivation of the oil with irgamet 39 is the most widely applied countermeasure to mitigate the negative effects of corrosive sulfur compounds, and also because of its relatively low-cost impacts [16]. Another alternative solution concerns the sulfur removal technique on-line and off-line: treatment with sorbents [17], selective depolarization [18], mobile reclaiming with reactivating sorbents [19], treatment with KOH/PEG [20] and liquid-liquid extraction [21]

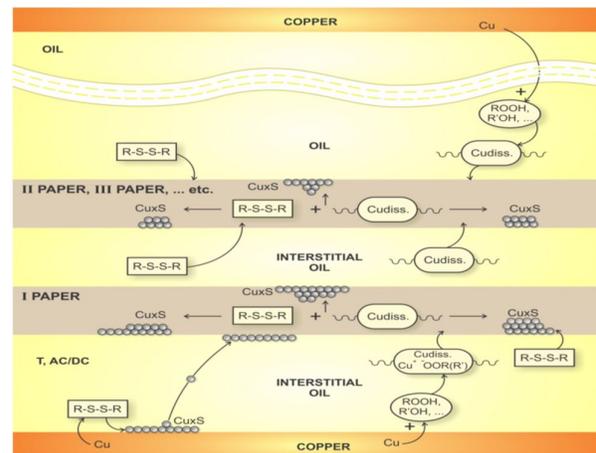


Figure-2. Proposed mechanism for the formation, migration and deposition of  $Cu_xS$ .

The replacement of the used oil with new ones is also a possibility. However, this technique is to be considered as a most extreme solution; it is generally used in very few circumstances, although considered by CIGRE



the most robust procedure based on the long-term stability of the properties of the insulating oil [2, 22].

### Design of experiment

The experimental design is widely used to optimize process parameter values in order to improve the quality of a product or a process. Full Factorial and One-Factor-at-the-time (OFTA) experiments are design methods that can possibly be used but require a large number of experiments when the number of process parameters increases. Taguchi developed the foundations of the Robust Design method introduced in the 1950s and 1960s and applied in electronics, automotive, photographic and many other industries, which has been an important factor in the rapid industrial growth of Japanese industries [23]. In science, the researcher's interest is focused on system (processes) investigations where, usually, there are several variables for analysis. Often, the investigations are centered on individual changes produced by the variables, as well in their interactions (Figure-3). In every process, there are several inputs and several outputs. The number of inputs or outputs can be one or more than one. Some of the inputs are selected as factors to be varied, and some outputs are selected as the responses to be measured and optimized. The combined study of multiple factors represents a way to determine the main effects, as well as the interaction effects among the factors underlying the process. The Design of Experiment (DOE) is a framework of statistical techniques, such as the results can produce valid and objective conclusions [24]. DoE is a scientific approach, which allows the researcher to better understand a process and to determine how the inputs affect the response [25]. DoE methodologies have been used very successfully in the verification, improvement and reducing the process variability with impacts on costs and development time [24]. A relevant class of DoE techniques is called factorial design, whose goal is to study and analyze the results (effects) produced by multiple variables of a process.

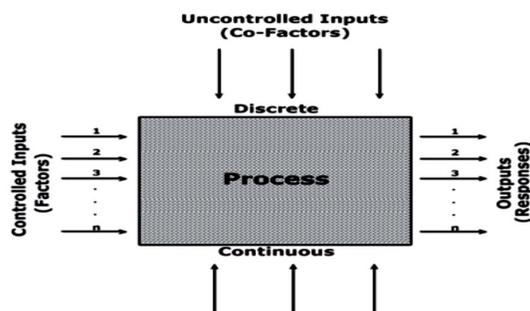


Figure-3. A classical process [24].

The beginning of a factorial design is a careful selection of a fixed number of levels for each set of factors. The experiments should be performed with all factor/level combinations. For example, if there are  $\ell_1$  levels associated to the first variable,  $\ell_2$  to the second, ...,  $\ell_k$  to the  $k$ -th factor, the full array of  $\ell_1, \ell_2, \dots, \ell_k$  plays

will be classified as factorial design  $\ell_1 \times \ell_2 \times \dots \times \ell_k$ . The default schema for designs with two levels uses the notation "-" (negative) and "+" (positive) to denote the low and high levels of each factor, respectively [24]. For example, a 2x2 factorial design with two factors  $X_1$  and  $X_2$ , and two levels (low and high), requires four experimental plays (Table-1).

Table-1. Two factorial design.

Experiment	$X_1$	$X_2$	Result
1	-	-	$Y_1$
2	+	-	$Y_2$
3	-	+	$Y_3$
4	+	+	$Y_4$

When all combinations of factors are running at the same number of times for each level, the experiment is classified as  $2^k$  full factorial design. The most intuitive approach to study such factors would be to vary the factors of interest in a full factorial design (trying all possible combinations of settings). For example, a  $2^3$  full factorial with three factors ( $X_1, X_2$ , and  $X_3$ ) at two levels would require eight experimental plays, while to study 5 factors at two levels, the number of runs would be  $2^5 = 32$ , and  $2^6 = 64$ , and so on. So, the number of runs required for  $2^k$  full factorial design grows geometrically as  $k$  increases. We can easily see that even if the number of factors is small, a full factorial design can become large quickly. In these circumstances, it is recommended [24] to use fractional factorial designs where only a fraction of appropriate combinations is required for execution of the full factorial design. Fractional designs are commonly used when one wants to investigate  $k$  factors with a smaller number of experiments, where  $p$  is the reduction factor [24].

### EXPERIMENTAL ARRANGEMENT

The materials used for the investigation are a 250 ml narrow-mouth glass, a copper foil (99 + % pure, 0.127 to 0.254 mm in thickness), and some 240-grit silicon carbide paper that serve as polishing material. The experimental procedure defined in ASTM Test Method D 1275 B [14] was used. With an abraded and polished copper strip (6 mm x 25 mm), 250 ml of non-filtered oil was poured into a flask having a glass stopper. The copper strip has to be bent in a V-shape before being added into the flask. The oil was then sparkled with dry nitrogen for 1 minute and sealed. The container was placed in an oven kept at 150°C, during a given a certain time period, to conduct the heating tests. The test container is shown in Figure-4. After the heating test, the copper strip was taken out from the test oil and rinsed in hexane. Then, the extent of its corrosion was measured.



**Figure-4.** Copper strip in oil.

### Impacts simulation

The effect of different parameters (oxygen, temperature and time) on our samples of copper was observed under various scenarios. First, the impacts of the three parameters were checked separately. Then, the combined impact of these parameters was simulated as they were put together. Since the transformers can operate either in a hermetically sealed or a configuration exposed to air, an important parameter to take into account is the presence and availability of oxygen [26]. Three types of oil (of the same brand) conditioned with 4259 ppm, 17169 ppm and 45018 ppm of oxygen content were used repeatedly. To reach the oxygen content, the oil samples were degassed. A gas chromatograph (GC-2014) manufactured by Shimadzu was used to determine the oxygen content. The selection of oxygen content was done knowing that the content of dissolved oxygen in an oil sample taken from the container of a freely breathing transformer ranges from 5,000 and 40,000 ppm [27]. The aging of the copper samples took 48 hours at 150°C. Only service-aged oil having 4259 ppm of oxygen was used. The aging of the three samples of copper lasted respectively 48 h, 96 h, and 168 h, at 150°C. The copper samples were aged at different temperatures (70°C, 115°C, 150°C) during 48 h. Service-aged oil sample with 4259 ppm of oxygen was used. These temperature levels include conditions beyond the maximum operating temperature allowed (according to IEC loading Guide, 105°C), and were chosen to accelerate the corrosion process. Specifically, the temperature of 115°C was chosen because it represents the expected hottest-spot temperature for a transformer with thermally upgraded insulation,

loaded and operating at full rated conditions [6]. The combined impacts are:

- combined effects of oxygen and time: Aging of the copper samples was based on different aging times (48 h, 96 h, 168 h) at 150°C. Oil was used with 45018 ppm of oxygen.
- combined effects of time and temperature: Oil with 45018 ppm of oxygen was used. Aging of the copper samples was done over different time periods and: (48 h, 96 h, 168 h) ⊗ (70°C, 115°C, 150°C). This crossing indicates that for each aging time, the sample of copper was submitted to different temperatures (70°C, 115°C, 150°C).
- combined effects of oxygen and temperature: Aging of the copper samples was based on different temperatures (70°C, 115°C, 150°C) during 48 h. Oil was used with 45018 ppm of oxygen.

### Determination of sulphur on the conductors

The overall sulphur analysis was conducted with the EMIA-220V induction furnace (Figure-5) from HORIBA, using the high-frequency furnace method, on the basis of infrared light absorption during combustion, in an oxygen flow. Some accelerators (Fe, Sn, W) were added to the sample, put into a porcelain crucible and heated in the induction furnace. Carbon transformed into CO<sub>2</sub> and CO, and sulphur into SO<sub>2</sub> when the sample reacted with the oxygen flow. The analysis showed that water was released (H<sub>2</sub>O or H<sub>2</sub>) and, since it is considered as a contaminant, it is eliminated by dehydration with magnesium perchlorate (Mg(ClO<sub>4</sub>)<sub>2</sub>). The oxygen flow is then regularized and passes through an infrared detector. A CO, CO<sub>2</sub> and SO<sub>2</sub> detector measures the concentration in sulphur.

### Statistical design

To optimize the conditions of the current process, we used an experimental design, which included a full factorial design with number of levels exceeding two. All three factors selected (Time, Oxygen and Temperature) as input variables were investigated at three different levels. The levels tested cover the range suggested in the literature. The levels chosen are given in Table-2.

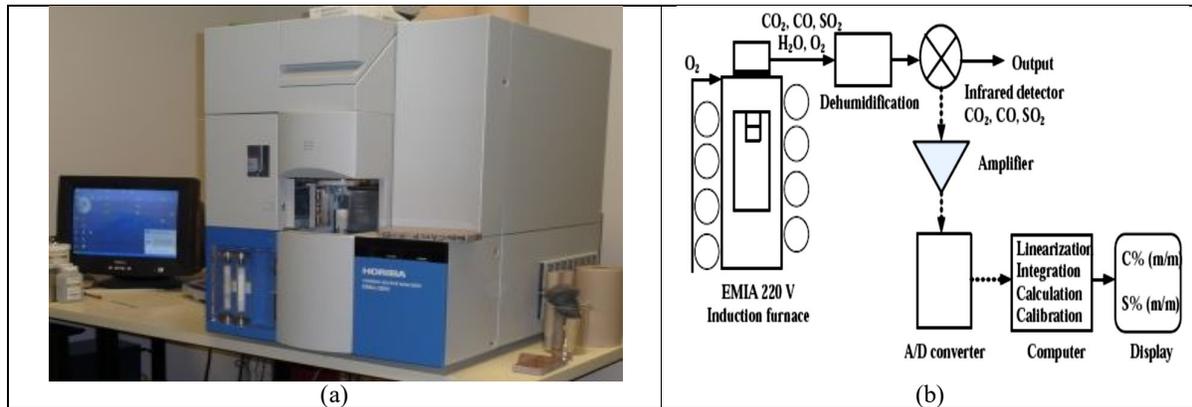


Figure-5. Experimental environment: a) Model EMIA 220 V induction furnace and b) the operating diagram.

Table-2. Factors and the corresponding levels considered in the DOE study.

Experiment N°	Factors			Response
	Time (h)	Oxygen (ppm)	Temperature (°C)	Sulfur (ppm)
1	48	4259	70	4,3
2	48	4259	115	7,9
3	48	4259	150	30,3
4	48	17169	150	41,5
5	48	45018	70	3,5
6	48	45018	115	12,7
7	48	45018	150	44,75
8	96	4259	150	75,7
9	96	45018	70	3,8
10	96	45018	115	14,7
11	96	45018	150	72,75
12	168	4259	150	146,7
13	168	45018	70	6
14	168	45018	115	24,5
15	168	45018	150	187,5

## RESULTS AND DISCUSSIONS

### Qualitative analyze of samples

Figure-6 shows the ASTM D-130 Copper Strip Corrosion Test [28] which color scale used to determine the tarnish level as a reference for the results. Detection of corrosive sulphur species is performed initially by the transformer oil manufacturer and then by the transformer operator. Although there are standard corrosion tests, these results are qualitatively analyzed. The test results are either pass or fail, based on the discoloration of the copper surface as compared to the standard color scale. Photos of copper strips under various impacts after heating test are shown in Tables-3 to 8 below. Tables 3-8 show the evolution of the corrosive sulfur indexing for the different impacts (oxygen, time, temperature) as represented by

changes in the color surface of copper conductors. The color of the copper strips turns gradually from yellow to light gray, dark gray and black. The presence of dark gray demonstrates that there is  $\text{Cu}_2\text{S}$  deposition according to [29]. The primary effect of the presence of corrosive sulfur species in insulating oil is the formation on copper sulfide ( $\text{Cu}_2\text{S}$ ) on the surface of copper conductors. The evolution of the sulfur index is highly variable. It shows the corrosiveness to copper of one sample only by the impact due to time. This copper sample corroded up to index 4a, for an oxygen content of 4259 ppm, a time of 168 hours and temperature of 150 °C. There was no copper corrosion as for the impact of temperature. Apparently, the impact of time is the most significant, followed by the impact of oxygen. The impact of temperature seems to be less aggressive. Surface discoloration of the copper specimens



demonstrates the growth of the copper sulfide films on the copper conductor specimens. Furthermore, the results suggest that the corrosion mechanism of different copper strips were not the same and that the corrosion ability, also varied in different environments.



**Figure-6.** ASTM D-130 color scale of the Copper Strip Corrosion Test.

**Table-3.** Results after the impact of oxygen.

	Oxygen @ 4259 ppm Time @ 48 h Temperature @ 150°C	Oxygen @ 17169 ppm Time @ 48 h Temperature @ 150°C	Oxygen @ 45019 ppm Time @ 48 h Temperature @ 150°C
Picture			
Tarnish level	2c	3b	3b
Results	Non-corrosive	Suspected corrosive	Suspected corrosive

**Table-4.** Results after the impact of time.

	Oxygen @ 4259 ppm Time @ 48 h Temperature @ 150°C	Oxygen @ 4259 ppm Time @ 96 h Temperature @ 150°C	Oxygen @ 4259 ppm Time @ 168 h Temperature @ 150°C
Picture			
Tarnish level	2c	3b	4a
Results	Non-corrosive	Suspected corrosive	corrosive

**Table-5.** Results after the impact of temperature.

	Oxygen @ 4259 ppm Time @ 48 h Temperature @ 70°C	Oxygen @ 4259 ppm Time @ 48 h Temperature @ 115°C	Oxygen @ 4259 ppm Time @ 48 h Temperature @ 150°C
Picture			
Tarnish level	1b	1b	2c
Results	Non-corrosive	Non-corrosive	Non-corrosive

**Table-6.** Results after impact of oxygen and time.

	Oxygen @ 45018 ppm Time @ 48 h Temperature @ 150°C	Oxygen @ 45018 ppm Time @ 96 h Temperature @ 150°C	Oxygen @ 45018 ppm Time @ 168 h Temperature @ 150°C
Picture			
Tarnish level	3b	4a	4c
Results	Suspected corrosive	Corrosive	Corrosive

**Table-7.** Results after oxygen and temperature.

	Oxygen @ 45018 ppm Time @ 48 h Temperature @ 70°C	Oxygen @ 45018 ppm Time @ 48 h Temperature @ 115°C	Oxygen @ 45018 ppm Time @ 48 h Temperature @ 150°C
Picture			
Tarnish level	1b	1b	3b
Results	Non-corrosive	Non-corrosive	Suspected corrosive

**Table-8.** Results after time and temperature.

	Oxygen @ 45018 ppm Time @ 48 h Temperature @ 70°C	Oxygen @ 45018 ppm Time @ 48 h Temperature @ 115°C	Oxygen @ 45018 ppm Time @ 48 h Temperature @ 150°C
Picture			
Tarnish level	1b	1b	3b
Results	Non-corrosive	Non-corrosive	Suspected corrosive
	Oxygen @ 45018 ppm Time @ 96 h Temperature @ 70°C	Oxygen @ 45018 ppm Time @ 96 h Temperature @ 115°C	Oxygen @ 45018 ppm Time @ 96 h Temperature @ 150°C
Picture			
Tarnish level	1b	1b	4a
Results	Non-corrosive	Non-corrosive	corrosive
	Oxygen @ 45018 ppm Time @ 168 h Temperature @ 70°C	Oxygen @ 45018 ppm Time @ 168 h Temperature @ 115°C	Oxygen @ 45018 ppm Time @ 168 h Temperature @ 150°C
Picture			
Tarnish level	1b	2c	4c
Results	Non-corrosive	Non-corrosive	corrosive

### Quantitative analyze of samples

The total level of sulphur in the new copper measured by the EMIA 220 V is 2.3 ppm [30]. Figure-7 shows the Sulphur content when impacts are considered independently. This Figure shows an increase in sulphur. The action of increased oxygen resulting in the increase in sulphur content shows how damaging oxygen is to power transformers. While at low temperatures (<115°C), the sulphur content is not alarming (since the sulphur amount is lower than the acceptable limit in copper ( $\approx 15$  ppm)), it becomes impactful from 150°C. It has been

experimentally shown that non-corrosive sulphur becomes corrosive when exposed to high temperatures [7]. The impact of time and oxygen results in high sulphur content. From this information, it is clear that the qualitative method is not sufficient to detect corrosive sulphur. The risk of copper corrosion can be confirmed with this value of sulphur content. Even with small amounts of oxygen (<5,000 ppm) as with transformer conservators (gas blanketed, sealed conservators) there is a formation of copper sulphide which agrees with the reported results [7, 31].

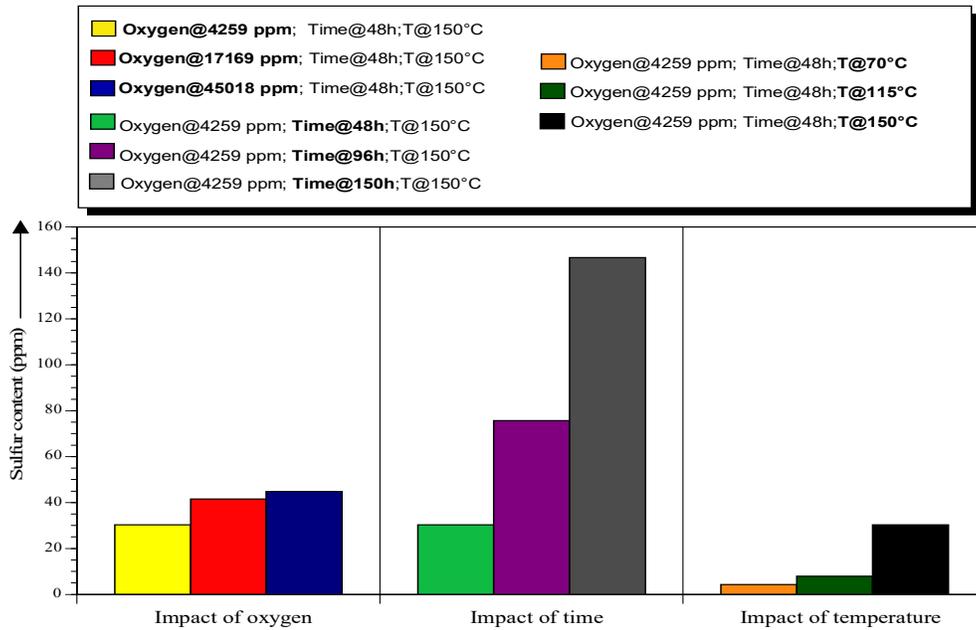


Figure-7. Impact of oxygen, time and temperature.

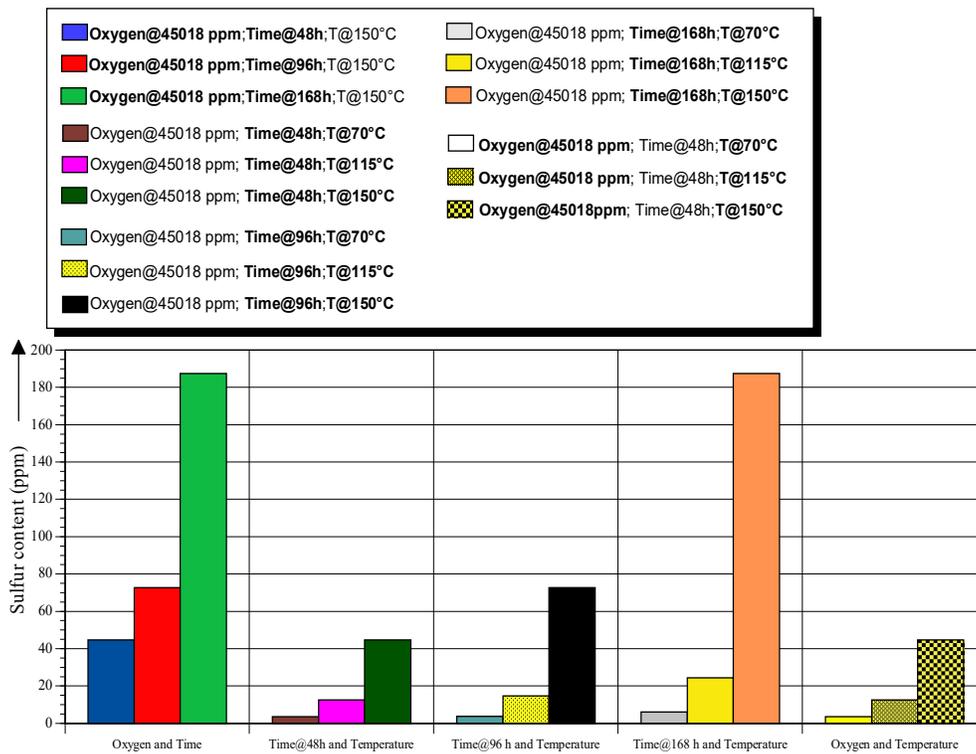


Figure-8. Comparison on the interactions between impacts

However, the qualitative method cannot diagnose it adequately. With the impact of time, the top sulphur content was 147.8 ppm which is roughly 65 times higher than its initial value. Recent works [3] have shown that the amount of Cu<sub>2</sub>S deposition on insulation paper and copper wire surface increases with aging time. Figure-8 shows the

sulphur content and a comparative recap when the impacts are considered together.

The above figure shows an increase of 187.5 ppm in the sulphur deposition. Note that the largest value reached is approximately 82 times higher than the initial value. Oxygen and sulphur have very similar properties and they are in the same group of elements. Over the last



decades, there has been a growing concern about the impact of oxygen on corrosive sulphur deposition. In the past, a low oxygen content of oil was believed to foster sulphur deposition [32]. This might be due to the low level of failures reported in free breathing transformers. From the result (Figures 7 and 8), it can be inferred that the combination of impacts shows a higher contribution in the production of the corrosive sulphur on the copper conductor. For a given amount of sulphur in oil, the combination of time and oxygen aggressiveness causes more damage. The impact of time and temperature is alike but only at high temperatures (from 150°C). At low temperature (70°C), the production of small quantities of sulphur points that copper is still usable. The combined effect of oxygen and temperature seems to be less harmful. In this case, the maximum value of sulphur deposition is 44.75 ppm, roughly 19 times the initial value.

### Results from experimental design

ANOVA (Analysis of Variance) is an important analysis tool for scientific experiments and it is also one of the most widely used statistical analysis methods. It is often used as a supplementary means of studying the variability of the means of experimental observations or to examine the significance of factors in a multi-factor experiment. All results in this study were generated by the statistical analysis commercial software Ellistat's DOE [33]. Pareto charts of DOE to discover the most influential parameters are showed in Figures 9 and 10. The Pareto charts depict that among the three parameters, temperature have the most influence on the formation of corrosive sulfur on the copper conductor. Oxygen is statistically not significant. When the impacts are partnered, the combination of time and temperature was found to have strong influence on corrosive sulfur.

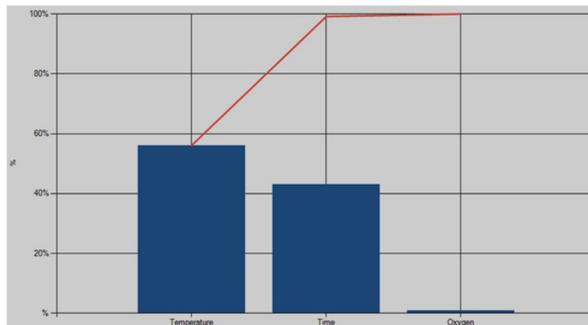


Figure-9. Pareto contributions to the variance in standardized ANOVA.

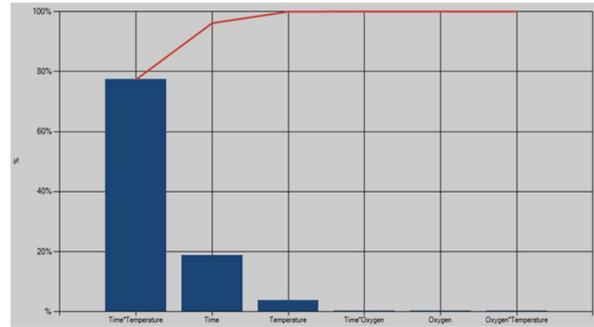


Figure-10. Pareto contributions to the variance in standardized ANOVA.

However, the combinations of time and oxygen, and oxygen and temperature have virtually no influence on the formation of corrosive sulfur. They are therefore not significant. Figure-11 shows whether the distribution is normally distributed and know the residual analysis. The residual value found in the experiment is  $p = 0.136$  according to the relevant Anderson-Darling test [24]. The results show that the hypothesis of normality is compatible with our data.

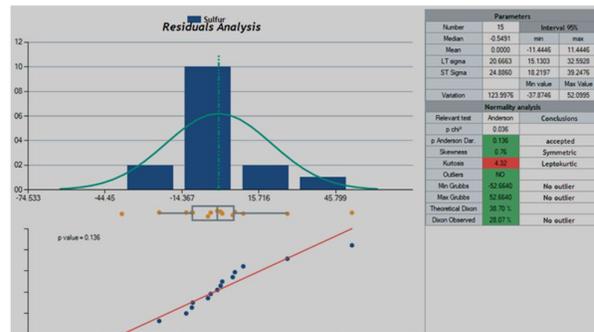


Figure-11. Residuals analysis.

### CONCLUSIONS

This study on deposition of corrosive sulfur on copper was conducted in accordance with the qualitative method ASTM D 1275 B and its experimental design included a full factorial design. The qualitative and quantitative effects of corrosive sulfur were analyzed under various scenarios. The obtained results show the necessity of determining the amount of sulfur contained in dissolved copper in oil. These results can be summarized as follows:

- ASTM D 1275 B is not sufficient by itself to deal with the issue of corrosive sulfur. While the qualitative method detects a non-corrosive copper, the quantitative method shows that this copper may be corrosive when the sulfur contained in the copper exceeds the permissible value of sulfur in the copper.
- The quantitative method allows diagnosing the parameters accelerating the formation of corrosive sulfur.



- Temperature is a nuisance parameter; it is first followed by the action of time and lastly by the action of oxygen.
- In oxygen deficient environments (< 5000 ppm), formation of corrosive sulfur may occur, but the qualitative method cannot detect it;
- The combined action of time and the aggressiveness of the dissolved oxygen produces high amounts of sulfur.
- The production of corrosive sulfur on a copper conductor is accelerated when temperature and time are partnered.

Identifying the sources of formation of corrosive sulfur should help to deal more efficiently with sulphur corrosion of copper whose effects are devastating, and which is a major problem for power transformers.

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