1 Distinct degradation processes in ZnO varistors: reliability analysis and modeling with accelerated AC tests 2 Hadi YADAVARI¹, Mustafa ALTUN^{1, *} 3 4 ¹Faculty of Electrical and Electronics, Istanbul Technical University, Istanbul, Turkey *Corresponding Author: altunmus@itu.edu.tr 5 Abstract: In this study, we investigate different degradation mechanisms of ZnO 6 7 varistors. We propose a model showing how the Vv (defined as the DC varistor voltage 8 when 1mA DC current applied) changes by time for different stress levels. For this 9 purpose, accelerated degradation tests are applied for different AC current levels; then voltage values are measured. Different from the common practice in the literature that 10 11 considers a degradation with only decreasing Vv values, we demonstrate either an increasing or a decreasing trend in the Vv parameter. The tests show us a decreasing trend 12 in Vv for current levels above a certain threshold and an increasing trend for current levels 13 below this threshold. Considering both of these degradation mechanisms, we present a 14 15 mathematical degradation model. The proposed model exploits physics of the 16 degradations for a single grain boundary that is the core structure of ZnO varistors. To validate the proposed model, we perform Monte Carlo simulations and the results are 17 compared with those obtained from accelerated AC tests. At the end as a summary of this 18 19 study, we introduce a conceptual accelerated AC test methodology to analyze the 20 reliability of a new ZnO varistor. Key words: Reliability analysis and modelling, degradation, ZnO varistors, AC tests. 21 22

23 1. Introduction

Zinc oxide (ZnO) varistors have been widely used in electrical and electronics systems 1 against overvoltage surges thanks to their non-ohmic current-voltage characteristics and 2 excellent energy handling capabilities [1, 2, 3]. ZnO varistors are variable resistors used 3 for limiting or diverting transient AC line voltage; they are usually subjected to long-term 4 5 AC voltage and surge stresses which lead to degradation of them. Even varistors used within their well-defined specifications might fail due to degradation; ageing 6 phenomenon makes the varistors degraded and even thermally broken down or destructed 7 8 [4]. Therefore degradation analysis of varistors is crucial.

9 Overwhelming majority of the studies in the literature have reported an increasing leakage current and accordingly an increasing Vv parameter (defined as the DC varistor voltage 10 11 when 1mA DC current applied), manifesting the degradation [1, 5, 6, 7, 8]. On the other hand, decreasing leakage current that results in an increase of Vv, is totally disregarded. 12 Only few studies have mentioned this without a detailed analysis [9, 10]. Here, the reason 13 is that as opposed to an increasing leakage current, a decreasing leakage current might 14 not hazardous for a varistor. However, we show and demonstrate that this kind of 15 16 degradation directly affect system reliability and safety [11]. This is illustrated in Figure 1. Even a slight increase in Vv leads to a dramatic change in maximum designed voltage 17 criteria for other components in the same block, especially for those in the power supply 18 19 block. This eventually causes a breakdown of the protected circuits and components. This 20 phenomena is demonstrated by testing over 100 electronic cards that were commercially used and failed due to varistor degradations [11]. As a result, to our knowledge, this is 21 22 the first study on modelling and characterization of ZnO varistors regarding both 23 degradation processes.

We perform accelerated AC voltage/current tests to analyze varistor degradations. In 1 general, accelerated tests are conducted by applying stresses such as temperature, 2 vibration, humidity, and voltage, beyond their normal/expected levels in field [12, 13]. 3 We use a single stress variable of voltage since it is by far the most dominant stress for 4 5 varistors. Indeed, there are two major reliability/degradation test methodologies for varistors that are pulse tests (8/20 µs and 2ms tests) and accelerated AC tests. We use 6 7 accelerated AC tests since it is cost efficient both in terms of test time and test equipment. 8 In the literature there are studies using AC test with showing that that the Vy parameter 9 of varistors which is one of the key parameters related to measuring reliability of a varistor, can be changed during the tests [7, 11, 14]. In this study, we make reasoning, 10 justification, and modelling of these Vv changes. By applying accelerated AC signals, we 11 see either an increasing or a decreasing trend in Vv that depends on the current levels 12 13 passing through the varistor. The tests show a continuous and fast decreasing trend in Vv for current levels above a certain threshold and a slow increasing trend for current levels 14 below the threshold. Although we apply accelerated AC tests to a particular metal-oxide 15 16 varistor -- detailed information such as composition, microstructure, and phases of this particular ZnO varistor is available in the data sheet [15], our results are generally 17 applicable to variety of similar metal-oxide varistors. 18

We investigate physical bases of the degradation processes observed in the AC tests. Physically, a ZnO varistor has a structure containing numerous number of zinc oxide grains of different shape and size with other metal oxide additives [15]. Several studies demonstrate that electrical stresses on varistors cause deformation of grain boundary potential barriers which are highly resistive [5, 16, 17]. It is reported that mild degradation after applying an electric field stress leads to a decrease in the effective doping

concentration of grains and accordingly an increase in the resistance of grain boundaries 1 [6]. On the other hand, applying a strong electrical field might lead to an increase in the 2 doping concentration resulting in a decrease in the resistance of grain boundaries [18]. 3 Motivated by this, in our model, we assume that every single ZnO-ZnO grain boundary 4 5 can be considered as a resistor. We formulate the resistor as a function of time and applied stress that determines relative effects of the degradation processes. Additionally, we need 6 7 to determine how to combine the resistors by considering grain shapes and amounts. In 8 this regard, several studies have been proposed aiming on varistor microstructures by 9 considering grain shape and topology as well as type and distribution of defects [19, 20, 21]. In a similar way, we present a simple yet accurate microstructure that solely consists 10 11 of grain boundary resistors. We perform a Monte Carlo method to validate our model; outcomes of the simulations match well with the results obtained from the accelerated 12 13 AC degradation tests.

The outline of paper is as follows. In Section 2, the proposed accelerated AC test methodology used to distinguish degradation processes is presented. In section 3, the physical bases of the processes are presented and used to form a varistor degradation model. Simulation results are also given to validate the proposed model. In the section 4, a reliability test methodology is presented to facilitate and summarize the steps proposed in this study. Section 5 reports the conclusion of this work.

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2. Degradation processes by accelerated AC tests

In order to investigate variator degradation mechanisms, we perform accelerated AC tests
which are cost efficient both in terms of test time and test equipment. The tests show
different degradation processes including stable, decreasing, and increasing trends in Vv

depending on the applied AC signal levels. Different accelerated AC signal levels are 1 applied on varistor samples having a diameter of 12mm and a height of 15mm as shown 2 in Figure 2. We number varistors starting from Var1 to Var11 which samples are of the 3 same material, manufacture, and characteristics [15]. 4 5 For a specific AC current passing through a varistor called as I_{VAR} (RMS value), voltage values of the varistor called as V_{VAR} (RMS value) are periodically measured. Our three-6 step test procedure is summarized as follows: 7 1) All samples are shorted to ground for 24 hours in order eliminate any previous 8 capacitive/inductive loads. 9 2) I_{VAR} is determined and continuously applied. Then V_{VAR} 's are periodically 10 measured for a 300 minute time period. If the varistor burns then the tests are 11 stopped at the time of the break down (failure). 12 13 3) Step-2 is repeated for different I_{VAR} values to analyze different degradation mechanisms and finally to find the IVAR threshold value. 14 For the second step, we need to determine which I_{VAR} values are used. Indeed, the 15 16 suggested I_{VAR} value according to IEC standard [IEC-60099-4] is 1mA [22]. However, we show that applying different I_{VAR} values results in different degradation mechanisms. 17 18 This is illustrated in Figure 3 with using 6 different I_{VAR} values 5mA, 3mA, 2mA, 1.5mA, 19 1mA, and 0.7mA. Note that for a certain I_{VAR} value, multiple samples are generally used 20 to take into account probable process variations. Analyzing the results in Figure 3, we certainly see two different degradation mechanisms corresponding to increasing and 21 22 decreasing V_{VAR} values over time. While a decrease in V_{VAR} results in an increase in leakage current and a decrease in Vv, increasing V_{VAR} makes the leakage current decrease 23 24 and Vv increase. Note that a varistor voltage Vv represents a varistor voltage when 1mA

DC current is conducted. Thus there is a positive correlation between V_{VAR} and Vv if a considerable amount of varistor current is conducted in the range of mA's. In the following section we separately analyze the cases in Figure 3 to find the threshold value of I_{VAR} that distinguishes between the degradation mechanisms.

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6 **<u>Finding the threshold:</u>**

In order to find the threshold value, we first tried $I_{VAR} = 10$ mA. Here, the varistor voltage 7 8 plunged dramatically and after seconds varistor started to burn up. We decreased this 9 value to 5mA but still there was a dramatic decline in the varistor voltage. Then we performed $I_{VAR} = 2mA$ tests for two samples as shown in Figure 4(a). Here, we do not 10 have not full data covering 300 minutes since the varistors started to burn up. We 11 12 continued reducing IVAR until VVAR values stabilized by time. This was achieved when 13 $I_{VAR} = 1.5$ mA. Indeed as shown in Figure 4(b), when $I_{VAR} = 1.5$ mA, V_{VAR} shows a slight decrease. It means that the threshold value is slightly under 1.5mA. We repeated the test 14 for $I_{VAR} = 1$ mA and $I_{VAR} = 0.7$ mA as shown in Figure 5 (a) and (b), respectively. 15

In conclusion, for current levels above 1.5 mA which is found the threshold value for this family of varistor samples, V_{VAR} drops dramatically until it burns. We classify this as a hard degradation mechanism. On the other hand, for current levels below the threshold value (1.5 mA), we see a relatively slow increasing trend in V_{VAR} . We classify this as a mild degradation mechanism.

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22 3. Physical bases of degradation processes and mathematical modeling

In this section, we aim to develop a mathematical reliability model for varistordegradations. For this purpose we first investigate physical bases of the degradation

processes observed in the AC tests. It is a general conception that electrical stresses on 1 varistors cause deformation of grain boundary potential barriers [5, 16, 17]. Figure 6 2 shows a conceptual microstructure of a ZnO varistor where grains and boundaries are 3 represented with white and grey regions, respectively. If boundaries are overcome then a 4 5 current is conducted through paths of grains that is illustrated with the arrows in the figure. Note that while ZnO grains are conductive, intergranular boundaries are highly 6 resistive. Since ZnO grains and boundaries are spread in almost a regular fashion, a single 7 8 grain boundary between two grains can be used as a core structure for varistor models. 9 This is indeed called a single grain boundary model [23, 24].

A ZnO-ZnO grain boundary can be modeled with a Schottky barrier as shown in Figure
7. By depletion of carriers from surrounding grains, a double Schottky barrier is forming.
Long-term stresses cause a degradation along with a change in the barrier height and
characteristics due to migration of ionized donors in electric field and redistribution of
them in the near-surface region of grains. This results in an increase in leakage current,
relevantly a decrease in Vv [25].

Other possible mechanisms of deformation of grain boundary barriers explain different processes of degradation in ZnO varistors. It is reported that there are two types of electron traps of importance for ZnO varistors: traps located at ZnO-ZnO grain boundaries known as interface traps, and traps located within the bulk of ZnO grains known as bulk traps. Interface trapping of electrons is generally considered as the mechanism giving rise to double Schottky barriers at grain boundaries [26].

Furthermore, it is reported that mild degradation after applying an electric field stress leads to a decrease in the effective doping concentration at the grains and accordingly an increase in the resistance of grain boundaries [6]. This increase clearly explains an increase in Vv. On the other hand, applying a strong electrical field might lead to an
 increase in the doping concentration resulting in a decrease in the resistance of grain
 boundaries [18]. Subsequently, it results in a decrease in Vv.

Motivated by the mentioned studies on physics of degradation demonstrating the change in grain boundary resistance values for different stress types, we model every single grain boundary barrier as a resistor. The resistor value can change according to applied stress in either increasing or decreasing trend. Thus, in this study, we contribute to a non-linear cross boundary resistor as a function of time and stress. This is illustrated in Figure 8.

9 The proposed model formula for a single resistor (grain boundary) is shown below:

$$R_t(t,s) = R_{in}(1 + \alpha R_i(t)R_i(s) + \beta R_d(t)R_d(s)).$$
(1)

11 Degradation depends on both stress (s) and time (t). The initial value of the resistor, degradation free value, is represented by R in that is obtained using initial V_{VAR} and I_{VAR} 12 13 values. Degradations mechanisms causing increasing Vv and decreasing Vv in time domain are represented by $R_i(t)$ and $R_d(t)$ functions, respectively. We select $R_i(t) = \exp(-\frac{1}{2})$ 14 nt^{1/m}) used for AC signal stresses. We select $R_d(t) = 1/\sqrt{t}$ that is derived using the leakage 15 current formula given in [16]. Although there is a common consensus in the literature for 16 decreasing Vv behavior in time domain shown by R_d (t), increasing Vv behavior and 17 corresponding physical models lack precision. Therefore, in determination of R_i(t), our 18 main source is the test data obtained from our experiments. The functions $R_i(s)$ and $R_d(s)$ 19 represent the effects of applied stresses on degradation. They could be in the form of the 20 following exponential/power functions [27]. 21

Arrhenius $R(s) = e^{c/s}$

Inverse power law $R(s) = s^n$

• Eyring $R(s) = s^{-1} e^{c/s}$

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Note that since the above equations are used for a single stress (s), they can be applicable for our case using $s = I_{VAR}$. In our accelerated tests we use a single stress of current I_{VAR} . According to the tests described in Section 2, the threshold current (I_{VAR-th}) is close to 1.5 mA that determines the effects and dominances of $R_i(s)$ and $R_d(s)$ for different I_{VAR} levels. Regarding this, we select $R_i(s) = (1-e^{-cs})$ and $R_d(s) = e^{c(s-IVAR-th)}$ where $s = I_{VAR}$. The coefficients in the degradation formula α , β , n, m, and c are empirically calculated using the test data. As a result: $\alpha = 0.003$, $\beta = 0.001$, n = 0.1, m = 0.7, and c = 4000.

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9 <u>Varistor microstructure formation and simulation results:</u>

Varistor ceramics are compound from ZnO grains of different shape, size, and orientation. 10 11 Since ZnO grains have predictable sizes, the total number of the grains in a varistor can be found with using the varistor size [15]. Suppose that each grain diameter is in the range 12 between 10 µm and 100 µm and a ZnO varistor has a size of nearly 5mm×10mm in two 13 dimensions [15] (the test samples) that results in 5 Thousand to 0.5 Million grains. Using 14 15 these relatively high number of grains or grain boundaries and corresponding non-linear 16 resistors in simulations is certainly unpractical, so we need a microstructure. Figure 9 shows a simplified varistor microstructure based on equal cubic grains or square grains 17 on a mesh which represents all grains and boundaries [19, 20]. We exploit this structure 18 19 by using resistors only for grain boundaries.

20 Considering the tested variator dimensions, we use X and 3X number of vertical and 21 horizontal squares, respectively. Here, increasing the value of X results in better accuracy 22 at the cost of worse runtime and complexity for the simulation. Therefore we need to 23 determine the minimum value of X for which we achieve relatively high accuracy. For 24 this purpose we start with X=1, and increase X one by one. It is apparent that for X=1,

there is no vertical grain boundary. For X=2 and X=3, corresponding microstructures are 1 shown in Figure 10. For each case we perform a Monte Carlo analysis using multiple 2 samples. This is illustrated in Figure 11. For all four graphs in the figure, we use the test 3 data obtained using $I_{VAR} = 2mA$. According to Figure 11, as expected increasing X or the 4 5 number of resistors improves the simulation accuracy that depends on the fluctuations between samples as well as the curve fitness to the test data. Analyzing the results in the 6 figure, we conclude that the cases X=3 (26 resistors) and X=4 (58 resistors) have almost 7 8 similar performances. Trying larger X values also give very similar results. Therefore we 9 decide to use X=3 with 58 resistors in further simulations.

Figure 12 shows the results comparing the test data obtained using I_{VAR} 's of 0.7mA, 1mA, 1.5mA, and 2mA and the related curves obtained using the proposed microstructure having 58 resistors. Note that since the varistor started to burn up after 130 minutes for 2mA current stress, there is limited test data for this stress level. Each of the 58 resistors is treated independently using the formula in (1). The results clearly proves the accuracy of the proposed model; there is almost a perfect match between the curves and the real test points.

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18 4. Reliability test methodology for a new varistor

A variety of endurance and environmental tests are conducted to assure the reliability of ZnO varistors. These tests are derived from the extremes of expected application conditions, with test conditions intensified to obtain authoritative results within a reasonable period [14]. The most commonly used test methodologies include "surge current derating (8/20 μ s)", "surge current derating (2 ms)", "fast temperature cycling", and "vibration" tests. Usually these methodologies are costly in terms of test time and test

equipment. Additionally, only the FAIL/PASS criteria is considered for most of these 1 methodologies, so they lack of analyzing reliability performance of varistors in time 2 domain. They also neglect different varistor degradation mechanisms explained 3 thoroughly in the previous sections. Considering these drawbacks, we present a simple 4 5 yet efficient test methodology to assess the reliability of a ZnO varistor based on accelerated AC degradation tests. Figure 13 shows a flowchart of the proposed 6 7 methodology. According to the flowchart, it is possible to analyze a varistor reliability 8 based on accelerated degradation AC tests with using the proposed degradation model.

9 First, accelerated AC tests are applied to find the threshold value that distinguishes between the degradation mechanisms. As examined in Section 2, the threshold value of 10 11 ZnO varistors should be close to a 1 mA current. Second, both heavy and moderate degradation tests are performed to specify the trend of the mechanisms. Third, the 12 proposed degradation model is used. The test data is fitted to the model by calculating 13 empirical coefficients from the experiments. By considering the obtained degradation 14 15 formula and the tested varistor usage in field, different critical degradation values can be chosen by designers. Finally, varistor reliability can be estimated. 16

Note that the proposed methodology is conceptually given without certain procedures and steps having specific numbers for different varistor families. These details are out of scope of this study and can be considered as a future work.

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21 **5.** Conclusion

In this paper, we study degradation processes for ZnO varistors. For this purpose, accelerated AC degradation tests are applied aiming on measuring varistor voltage values in time domain. As opposed to the common practice in the literature that considers a degradation with only decreasing Vv values, the tests show either an increasing or a
decreasing trend in the Vv parameter. To justify the observed degradation processes,
physical bases of the degradations are investigated. For this purpose, a single grain
boundary (Schottky barrier) is modeled as a non-linear resistor; its characteristics in time
domain changes with applied stress levels. Then a microstructure is formed using these
boundary resistors.

We perform a Monte Carlo method to validate the proposed varistor degradation model;
outcomes of the simulations match well with the results obtained from the accelerated
AC degradation tests. As a summary of this study, we introduce a conceptual accelerated
AC test methodology to make the reliability analysis of a new ZnO varistor.

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- 2 Figure 1. Breakdown of the system in case of different degradation processes of ZnO
- 3 varistor[11].



- **Figure 2.** Varistor samples used in the accelerated AC tests.



2 Figure 3. Accelerated degradation tests results: V_{VAR} values in time domain for different

- $3 I_{VAR}$ values.
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Figure 4. Accelerated degradation tests results: V_{VAR} values in time domain for (a) I_{VAR}
= 2mA and (b) values I_{VAR} = 1.5mA.



2 Figure 5. Accelerated degradation tests results: V_{VAR} values in time domain for (a) I_{VAR}

3 = 1mA and (b) values $I_{VAR} = 0.7$ mA.



Figure 6. Microstructure of a varistor element [15].





Figure 7. A simple double Schottky barrier model of a ZnO-ZnO grain boundary.





Figure 8. Modeling of a single grain boundary as a resistor.





8 Figure 10. Proposed microstructures (a) X=2, 6 resistors and (b) X=3, 26 resistors.



2 Figure 11. Simulation results in comparison with the AC test data for (a) single resistor,

3 (b) X=2, 6 resistors, (c) X=3, 26 resistors, and (d) X=4, 58 resistors.



2 Figure 12. Accelerated degradation test data in comparison with the simulation results

3 for (a) $I_{VAR} = 0.7mA$, (b) $I_{VAR} = 1mA$, (c) $I_{VAR} = 1.5mA$, and (d) $I_{VAR} = 2mA$ current

4 stresses.





Figure 13. Reliability test methodology flowchart for ZnO varistors.