Josef Pihera, Václav Mentlík, Radek Polanský
University of West Bohemia, Pilzno

THE ELECTRIC PROPERTIES OF CURED MICA-GLASS COMPOSITES

WŁAŚCIWOŚCI DIELEKTRYKÓW OPARTYCH NA KOMPONENTACH ŻYWICA/MIKA/SZKŁO

Abstract: The curing of Resin-rich composites materials is an important part of technology of electrical rotating machines main wall insulation manufacturing. The using of optimal curing temperature increases the service reliability of insulating system as well as the whole electrical machine. The diagnostics of changing properties during the curing and the estimation of the optimal curing temperature are thus important components at the manufacturing of insulating system of electrical machines.

The paper describes the behaviour of diagnostic parameter tan δ of cured mica-composites in dependence on curing temperature.

The studied material consists of two different three-layer mica composites, which are normally used for the manufacture of main wall insulation in electrical rotating machines. The basic components of the composites are: mica paper, epoxy-novolac resin and glass fabric. The particular variants of composites differ each from other in the type of curing agent. The specimens were cured over six different temperatures in the range from 130 to 180 °C. The main diagnostic parameter tan δ was measured online during the curing (curing characteristics) and after the curing process. The temperature dependence of tan δ was measured after the isothermal curing.

An analysis of the results allows us to determine the optimal curing temperature where the value of tan δ of the cured system is the lowest of all and the properties of temperature dependence of tan δ are the most suitable.

The magnitude tan δ gives very good information capability about materials during their curing and enables to determine the optimal curing temperature. The tan δ is also able to determine possible undercuring of the insulation, which occurs at lower curing temperatures.

Streszczenie: W celu zapewnienia wysokiej niezawodności systemów elektroizolacyjnych wirujących maszyn elektrycznych należy zwracać szczególną uwagę na prawidłowość przebiegu procesu technologicznego izolowania, impregnowania i utwardzania układu elektroizolacyjnego. Właściwe utwardzanie układu Resin-rich jest jedną z najważniejszych operacji podczas produkcji układu elektroizolacyjnego. W tym czasie następuje wzajemne przenikanie wiązań żywic i układ uzyskuje końcowe właściwości izolacyjne. Ustalenie optymalnej temperatury i czasu utwardzania jest niezwykle ważne. Opracowana została metoda określania właściwych czasów i temperatur utwardzania. Metoda badań (zwana charakterystyką utwardzania), pozwala obserwować zakres zmian współczynnika stratności tan δ podczas procesu utwardzania (w niniejszym artykule zawarto wyniki obserwowane dla warunków izotermicznych). Pomiary zależności współczynnika tan δ od temperatury wykonano w celu uzyskania bardziej szczegółowych informacji o właściwościach systemu izolacyjnego. Ta analiza pozwoliła stwierdzić, iż na skutek niewłaściwej temperatury utwardzania, mogą pojawić się niewłaściwe parametry układu izolacyjnego. Opisywana metoda badawcza może być stosowana do określania parametrów nowego rodzaju układu elektroizolacyjnego lub do modernizacji i rozwoju już istniejących materiałów, w celu osiągnięcia zmiany ich parametrów funkcjonalnych dla produkowanych maszyn elektrycznych.

W artykule przedstawiono mechanizm przemian zachodzących w dielektryku podczas utwardzania. W rozdziale 2 omówiono teoretyczne zależności, dotyczące współczynnika przenikalności elektrycznej ε^*, zawierającego zarówno część rzeczywistą (ε'), jak i urojoną (ε''). Omówiono tu równanie Havrilak-Negami (4), opisujące zmiany współczynnika zespolonej przenikalności.

Przygotowano eksperyment badawczy dla dwóch nowych typów materiałów Resin-rich. Realizowano utwardzanie płaskich próbek dla sześciu izotermalnych warunków procesu technologicznego, od 130 do 180 stopni C. W maszynach elektrycznych wirujących izolację główną tworzą materiały mika-szkło, zawierające papier mikowy, włókno szklane i żywicę epoksydową. W przeprowadzanych testach zastosowano żywice A i B, różniące się między sobą zawartością utwardzacza. Ocenę materiału elektroizolacyjnego przeprowadzono mierząc charakterystyki współczynnika tan δ, jak również zależności tan δ od temperatury (tab. I).

W tabeli II zawarto obserwowane wartości współczynnika tan δ w funkcji temperatury utwardzania po 200 minutach utwardzania. Rysunki 1 i 2 przedstawiają ww. charakterystyki w formie graficznej. Jako optymalną

temperaturę utwardzania przyjęto ok. 150 stopni C z uwagi na to, iż współczynnik tan δ przyjmuje wtedy najniższe wartości dla ustalonego czasu wygrzewania. W celu zweryfikowania otrzymanych rezultatów, przeprowadzono pomiary zależności wartości współczynnika tan δ od temperatury (dla materiałów utwardzanych przez 200 minut). Wyniki tych prób przedstawiono na rys. 3 i 4. Potwierdzają one fakt, iż optymalną temperaturą utwardzania jest temperatura 150^{O}C.

Przeprowadzone badania zmian właściwości dielektryków opartych na komponentach żywica/mika/szkło pozwoliły określić optymalną temperaturę utwardzania w celu zminimalizowania strat dielektrycznych układu elektroizolacyjnego. W celu określania optymalnych temperatur utwardzania można stosować jedną z metod analizy strukturalnej (np. DTA) [9]. Autorzy dziękują za wsparcie i pomoc Ministerstwa Edukacji Młodzieży i Sportu Republiki Czeskiej przy realizacji badań.

1. Introduction

The proper technological steps are required by manufacturing of main insulation system of rotating machines due to the service reliability assurance. The curing of Resin-rich insulating materials is one of the most necessary technological operations. The crosslinking of bond resins occurs at this process and the insulating material acquires the final insulating properties. The assessment of optimal curing time and temperature when the system is properly cured is very important. The suitable test method for determination of mentioned parameters was developed. The test method (called curing characteristics) continually observes magnitude of tan δ during the curing process (in the case of presented paper under isothermal conditions). The measuring of tan δ in dependence on temperature was realized for more detailed information about insulating system properties. This analysis discovers the possible inadequacies in cured material due to the improper curing temperature. The mentioned methods are suitable for properties evaluation of new developed materials during their application test as well as for evaluation of already existing materials during the manufacturing of new insulating system of electrical rotating machines.

2. Theory

The behavior of a dielectric during the curing is primary affected by polarization and conduction mechanisms. Complex permitivity ε* consists of a real (ε′) and imaginary (ε′′) part

$$\varepsilon^* = \varepsilon' - j\varepsilon'' \tag{1}$$

The real part of ε* is equal to the relative permitivity of the dielectric. The loss factor ε′′ is proportional to losses and it is equal to energy losses due to dipole movement under an ac electrical field. The loss factor ε′′ is given by the equation:

$$\varepsilon'' = \varepsilon''_{ion} + \varepsilon''_{dipol} \tag{2}$$

The subscript ion indicates the contribution of the ionic conductivity while the subscript dipole indicates the contribution of dipolar relaxation to the loss factor ε′′.

The part ε''_{ion} is given as:

$$\varepsilon''_{ion} = \frac{\sigma}{2\pi f \varepsilon_0} \tag{3}$$

where σ is the ionic conductivity, f is applied frequency in Hz and ε0 is the dielectric constant of vacuum.

At the beginning of the curing reaction (low degree of cure), the contribution of ionic conductivity is dominant and the value of relative permitivity ε′ is high [3]. The epoxy crosslinking and dipole relaxation become more dominant with further proceeding of curing reactions [4], [7]. This can be monitored as a peak in the dielectric dissipation factor. The contribution of relaxation is dominant at this moment and overlaps the contribution of ionic conductivity.

The Havriliak-Negami function (4) describes this behaviour of complex permitivity, loss factor and dipolar contribution respectively.

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{\left[1 + (j\omega\tau)^\alpha\right]^\beta} \tag{4}$$

where τ is relaxation time, ω is applied frequency, ε_0 is relaxed or static dielectric permitivity, ε_∞ is the unrelaxed permitivity, α,β are parameters between 0 and 1. The exponent α describes the width of the distribution of relaxation times while the exponent β describes the skew of the distribution of relaxation times.

Eq. (4) leads to: [4]

$$\varepsilon' = \varepsilon_\infty + \frac{\Delta\varepsilon \cdot \cos\beta \cdot \zeta}{\left[1 + 2(\omega\tau)^\alpha \cos\left(\frac{\alpha\pi}{2}\right) + (\omega\tau)^{2\alpha}\right]^{\frac{\beta}{2}}} \tag{5}$$

$$\varepsilon'' = \frac{\Delta\varepsilon \cdot \sin\beta \cdot \zeta}{\left[1 + 2(\omega\tau)^\alpha \cos\left(\frac{\alpha\pi}{2}\right) + (\omega\tau)^{2\alpha}\right]^{\frac{\beta}{2}}} \tag{6}$$

$$tg\zeta = \frac{(\omega\tau)^\alpha \cdot \sin\left(\frac{\alpha\pi}{2}\right)}{\left[1 + (\omega\tau)^\alpha \cos\left(\frac{\alpha\pi}{2}\right)\right]} \tag{7}$$

where $\Delta\varepsilon = \varepsilon_0 - \varepsilon_\infty$

It is possible to study the parts of complex permitivity during the curing with application of this analysis.

3. Experiment

The experiment concerning about the curing of two different Resin-rich materials was prepared because of new material development and its application test necessity. The curing of flat samples was realized under six different isothermal temperatures in the range from 130 to 180 °C.

The mica-glass composites usually used as main wall insulation of rotating machines contains mica paper, epoxy resin and glass fiber. The particular variants of composites A, B (used in the experiment) differs from each other in the type of used curing agent of epoxy resin. Evaluation of the materials was performed with using of curing characteristics tan δ measurement as well as with the measurement of tan δ temperature dependence. The temperature dependence of tan δ was performed on samples cured for 200 min. (from 40°C to 180°C with 1°C temperature increasing.)

Tab. I. Order of experiments

	Curing temperature / °C 130, 140, 150, 160, 170, 180			
	Curing time / min			
Used Analysis	10	30	60	200
Curing characteristics of tan δ	+	+	+	+
tan δ temperature dependence				+

4. Results and discussions

Described experiment discovers different curing properties and tan δ magnitudes at different curing temperatures. Table 2 presents the dependence on curing temperature.

Table II.
Values of tan δ depending on curing temperature after 200 min of curing

Curing temperature / °C	Material A	Material B
130	0,309	0,065
140	0,278	0,064
150	0,193	0,063
160	0,279	0,101
170	0,253	0,140
180	0,279	0,254

Figures 1, 2 present curing characteristics of tan δ in dependence on curing temperature. The peak of tan δ (given by dipolar relaxation [8]) depends on curing temperature - the higher the curing temperature, the earlier the peak appears (due to faster curing at higher temperatures)

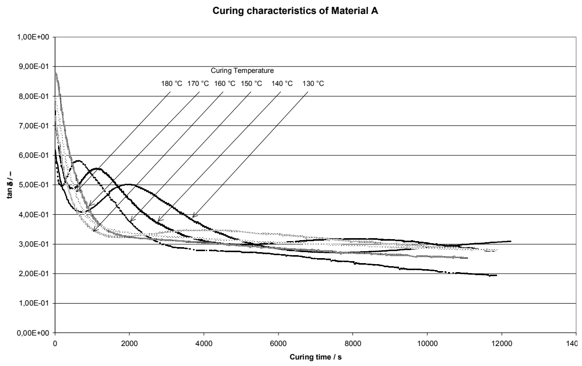

Fig.1. Curing characteristics of material A

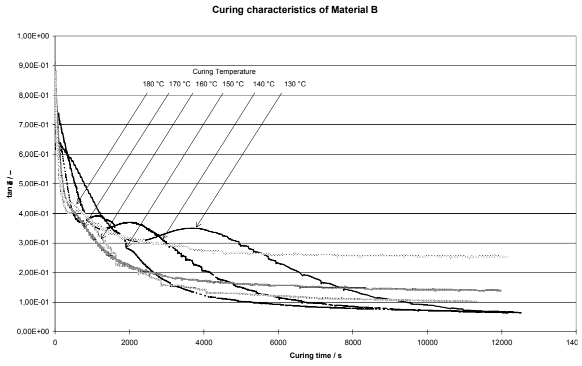

Fig.2. Curing characteristics of material B

At low curing temperatures material B does not show increased final dielectric loss values unlike material A which shows a strong upward trend, however at high temperatures >160°C both materials show increased dielectric loss factors. This fact should be theoretically explained by two hypotheses. First, the delamination of composite due to high temperature and consequently the beginning of micro-inhomogeneities leads to the loss factor increases. The second hypothesis of increased tan δ is possible rise of polar particles due to isomerisation processes inside the material structure [1]. The optimal curing temperature of 150°C is estimated from curing characteristics. The tan δ magnitude of cured material reaches the lowest value at this curing temperature.

The measurement of tan δ temperature dependence (at materials cured for 200 min.) was proceeded for verification of these results.

Figures 3 and 4 present experimental data of this test.

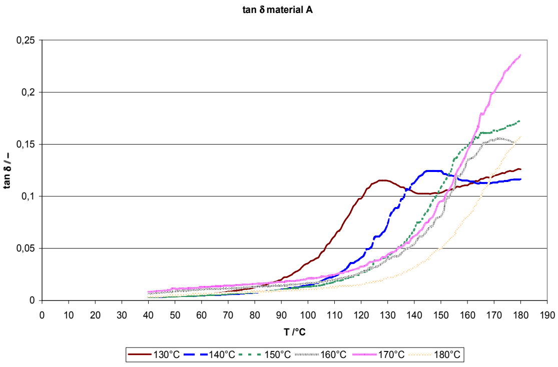

Fig.3. Temperature dependence of tan δ of material A

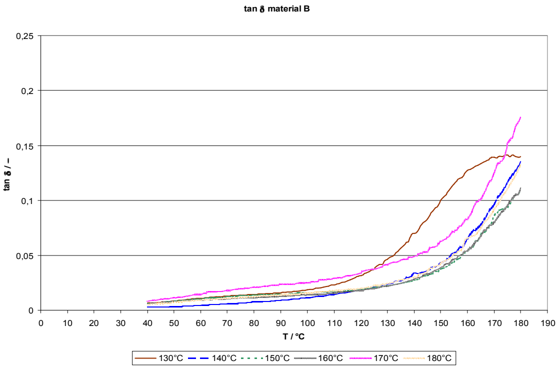

Fig.4. Temperature dependence of tan δ of material B

The imperfect curing of material A is evident at curing temperatures of 130°C and 140°C. The decreasing of tan δ comes into effect at temperatures near to the curing temperature. This is because of further curing of epoxy resin.

Study of mutual relationships between curing characteristics and tan δ temperature dependence confirms the most suitable curing temperature 150°C. The optimal electrical properties are evident from presented figures.

5. Conclusions

A detailed investigation shows the behaviour of the dielectric properties during the curing of epoxy/mica/glass composite systems A, B. This paper has described the differences in behaviour of two composite materials during the curing and has shown a method of determining the optimal curing temperature in order to minimize the dielectric loss of the cured system. The optimal curing temperature of the current materials appears to be 150 °C.

It is suitable to use any structural analysis (for example DTA) for the determination of optimal curing time [9].

The DTA test method, which follows the rate of reaction-able particles within material, allows to determine the moment of fully cured resin [1]. Thanks to this application the undercuring doesn't occur.

6. Acknowledgement

This article was carried out with the help of Ministry of Education, Youth and Sports of Czech Republic, MSM 4977751310 – Diagnostic of Interactive Processes in Electrical Engineering.

7. References

[1]. V. Mentlík, : *Experience with the Application of Thermal Analysis in High-Voltage Insulation*, Journal of Thermal Analysis, vol. 39/1993, pp. 1355

[2]. Pihera, J.: *New Materials for Electrical Engineering.* Dissertation thesis, Pilsen (Czech Republic) : University of West Bohemia 2005

[3]. Eloundou, JP. : *Dipolar relaxations in an epoxy-amine system*, European Polymer Jounal 38 (2002) 431-438

[4]. Lane, J. W.; Seferis, J, C. Polym. Eng. Sci 1986, 26, 346

[5]. IEC 250 Standard : *Recommended methods for determination of the permitivity and dielectric dissipation factor of electrical insulating materials at power, audio and radio frequencies including metre wavelengths*

[6]. Hongkyeong, K. : *Dielectric Changes During the Curing of Epoxy Resin Based on the Diglycidyl Ether of Bisphenol A (DGEBA) with Diamine*, Bull. Korean Chem. Soc. 1999, Vol. 20, No. 11

[7]. Montserrat, S. : *Vitrification and dielectric relaxation during the isothermal curing of an epox – amine system*, Polymer 44 (2003) 101-114
[8]. Ye-shiu Li.: *Kinetics and Curing Mechanism of Epoxy and Boron Trifluoride Monoethyl Amine Complex System*. Journal of Polymer Science: Part A: Polymer Chemistry, Volume. 37, 1999, 3614-3624
[9]. Mentlik, V.: *Material enthalpy – the classifier of curing of dielectrics containing epoxy resins*, IEEE CEIDP, Nashville, USA, 2005 pp.597-600

Autors

Pihera Josef, Mentlík Václav, Polanský Radek
University of West Bohemia, Faculty of Electrical Engineering, Department of Technologies and Measurement

pihera@ket.zcu.cz, tel. 00420377634520