

# Preparation and Optimization of Poly(Vinyl Formal) from Poly(Vinyl Acetate) for Electrical Insulation

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**Abstract**— The preparation of poly(vinyl formal) suitably used for wire insulation from commercially available local substances was investigated in this study. Such resin was prepared with higher formal content up to 82.3% from poly(vinyl acetate), acetic acid, and formaldehyde by solution polymerization technique which consists of hydrolysis of poly(vinyl acetate) to poly(vinyl alcohol) and this subsequently converted by formalization to poly(vinyl formal) at 70°C and reaction time of 24 hours. The three grades are 29, 35, and 50% of poly(vinyl acetate) have been investigated, only poly(vinyl acetate) with 29% solid yields a polymer nearest to that reported in the literature, also different amount of glacial acetic acid were used. Hydrolysis reaction seems highly enhanced by the presence of glacial acetic acid as solvent. The prepared poly(vinyl formal) was mixed with resole phenolic resin to produce coating and improve the mechanical, electrical, and thermal properties of the poly(vinyl formal). Different concentrations, temperatures, and curing time periods and were examined to find the optimum conditions: concentration of 10wt% resole phenolic resin, 162°C curing temperature, and 30 min curing time.

**Keywords**— Poly(vinyl formal), poly(vinyl acetate), electrical insulation, mechanical, electrical and thermal properties

## I. INTRODUCTION

Poly(vinyl formal) is part of polymer group termed as poly(vinyl acetal), that the first reported poly(vinyl acetal), poly(vinyl benzal) was prepared in Germany in 1924 by the reaction of benzaldehyde with poly(vinyl alcohol) in the presence of mineral acid catalyst.

Poly(vinyl acetal)s were commercialized during the 1930 and 1940 following development work at a number of firms including Dupont, Monsanto, Union Carbide and other in the United states and Canada .

In 1953 only poly(vinyl acetal)s commercially available are poly(vinyl formal) and poly(vinyl butyral), they were prepared by the acid catalyzed acetalization reaction of poly(vinyl alcohol) with aliphatic aldehydes. Formaldehyde and butyraldehyde were the only aldehydes which were commercially used to prepare poly(vinyl formal) (Formvar) and poly(vinyl butyral) [1].

Synthesis of poly(vinyl formal) consists of two sequential reactions. Poly(vinyl acetate) is first hydrolyzed to poly(vinyl alcohol) which is then reacted with formaldehyde to form poly(vinyl formal).

Poly(vinyl formal) is mainly used in three general applications wire coatings, surface coatings and adhesives.

The first and extensive application of poly(vinyl formal) is in the manufacture of a tough, adhesive and heat resistant wire enamel. In this application a standard enamel is based on a blend of low acetate poly(vinyl formal) with alkyl phenolic resin, after application to the wire this coating is dried and cured at a high temperature to a cross-linked film which is extremely tough and heat resistant. Such properties enable the magnet wire to maintain its coating integrity during high speed motor winding and service [2].

This work aims to prepare a suitable polymer (poly vinyl formal) from commercial available substance and subject it to various tests to examine its electrical, mechanical and thermal properties. The properties of poly(vinyl formal) were manipulated through blending it with suitable resin (alkyl phenolic resin) at a different concentration of each of them and at different curing conditions (temperature and time).

## II. MATERIALS AND METHODS

### A. Raw Materials

The materials used for poly(vinyl formal) preparation are: Poly (vinyl acetate), Glacial Acetic acid, Sulfuric acid , and Formaldehyde available at purities 29, 99-100, 97, & 37-40% respectively. The poly(vinyl acetate) was provided by the Chemical Engineering Department/ Baghdad University, Iraq & the other chemicals by Riedel- Dehgen, Germany.

### B. Experimental Procedure

The Poly(vinyl formal) was prepared by solution polymerization process involving hydrolysis of 345 g of PVA and formalization in a mixture of 500 g glacial acid and 35 distilled water in a three-necked round bottom flask. the resulted mixture was then heated up in a water bath type D1-91126 Schwabach-Memmert; Germany with temperature set at 70°C with maintaining constant stirring for one hour until the PVA is completely dissolved.

Then 4 g of 0.5M H<sub>2</sub>SO<sub>4</sub> and 60 g formaldehyde were added to the prepared solution and the reaction was allowed to take place for 24 h to get the desired composition of the final polymer product.

After 24 hour period, water was added to the mixture with rapid agitation in order to get a precipitate of the granules which was then washed many times with distilled water to remove the excess acid that may exist in the product; the acidity were measured by litmus paper and washing was continuous until neutralized product was obtained; meanwhile precipitation and filtration of poly (vinyl formal) granules was performed by Buchner funnel under vacuum; the final washed product was collected and dried by warm air at 30°C in a dryer. Figure 1 shows the vessel used for production of poly(vinyl formal).

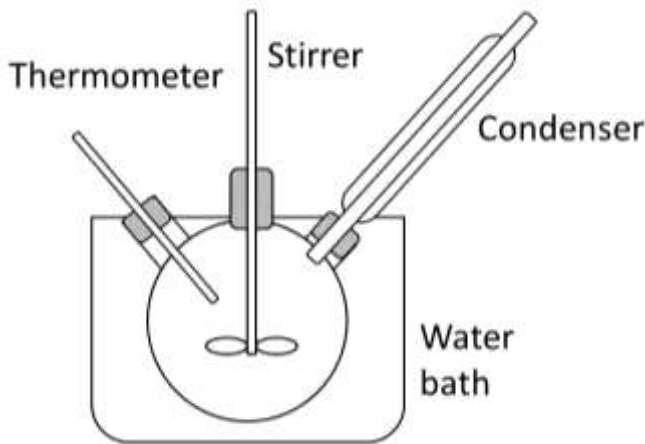


Fig. 1 Schematic diagram of the vessel used for poly(vinyl formal) preparation

C. Preparation of Coating Materials

Preparation of coating materials can be divided into two main stages: Dissolving poly(vinyl formal) in specific quantity of solvent (acetic acid) and mixing with Alkyl phenolic resin that consists of 80% solid [3] which is shown to give a suitable coating properties; reaction was performed at room temperature.

In order to obtain the best condition at which poly(vinyl formal) mixed with resole phenolic resin, three variables were investigated which are: Concentration of resole phenolic resin in mixture of poly(vinyl formal)-resole phenolic resin with the range of 10-90wt%, curing conditions temperature ranging between 100 and 225°C and time range of 10-30 min.

III. RESULTS AND DISCUSSION

A. Preparation of Poly(vinyl formal)

The poly(vinyl formal) obtained according to the procedure explained in Section II.B was identified using Infrared Spectroscopy Type Sp3-100-Sp-300 Spectrophotometer, England.

In this study three grades of poly(vinyl acetate) were used 50, 35, & 29% solid content: the samples 50% and 35% solid have failed both to give the desired product, only poly(vinyl

acetate) with 29% solid yielded a precipitated polymer in the form of a spongy granules.

The effects of acetic acid on final compositions (hydroxyl, acetate and formal) are shown in Table 1

The polymer prepared with compositions of hydroxyl, acetate and formal was found to be similar to that suggested by Saunders [4] for preparation of poly(vinyl formal) where the final product consists of 5 – 6 wt% hydroxyl and 10-13wt.% acetate in order to react with resole phenolic resin to prepare wire coating.

TABLE I  
COMPOSITION AS WT.% OF POLY (VINYL FORMAL)

Hydroxyl content %	Acetate content %	Formal content %
5.3	12.4	82.3

B. Physical Properties

The density and electrical conductivity of the liquid mixture poly(vinyl formal)-resole phenolic resin were determined before curing and coating at room temperature, as shown in Figure 2.

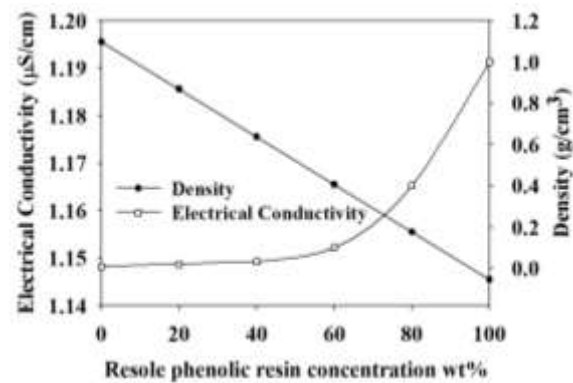


Fig. 2 Electrical conductivity in µS/cm and density of mixture poly(vinyl formal)-resole phenolic resin against concentration of resole phenolic resin

C. Mechanical Properties

The shear stress, elongation and peeling stress have been tested and plotted against the resole phenolic resin concentration wt%, curing time and curing temperature as shown in Figures 3, 4, and 5, which show that increasing phenolic resin will decrease the mechanical properties and the reduction of curing time will also decrease the mechanical properties, which is attributed to that fact that poly(vinyl formal) may act as plasticizing material while resole phenolic resin acts as a less plasticizing material and almost acts as a rigid material with no elasticity [5]. Also it indicates that high resole phenolic resin concentration will reduce all mechanical properties.

As for the curing time, its reduction to 20, 14.2 and 10 min, will compromise the mechanical properties greatly because the crosslinking through the ether linkage formed between

methylol group of the resole phenolic resin and the hydroxyl one belongs to the poly(vinyl formal) will not improve to the degree of high curing time.

In Figure 6, 7, and 8 the effect of curing temperature is shown such that increasing resole phenolic resin concentration to 162°C was found to improve all mechanical properties. Increasing the temperature beyond 162°C had the mechanical properties to decline up to 225°C where the sample failed the elongation test. This may be due to the fact that increasing curing temperature will cause an increase in the crosslinking through the ether linkage formed between methylol group of the resole phenolic resin and the hydroxyl one of the poly(vinyl formal) until the certain temperature is reached so that the crosslinking is completed at 162°C. That increasing curing temperature will cause a material to lose its elasticity and become very rigid which will lead to the failure of the material when subjected to high stress.

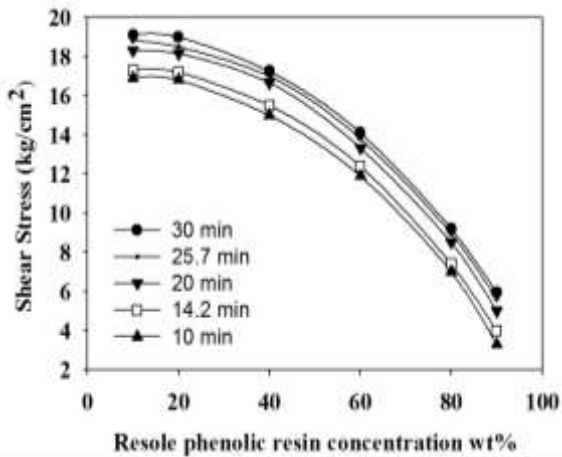


Fig. 3 Shear stress vs. phenolic resin concentration at different curing time from 10 to 30 min and at fixed curing temperature of 162°C

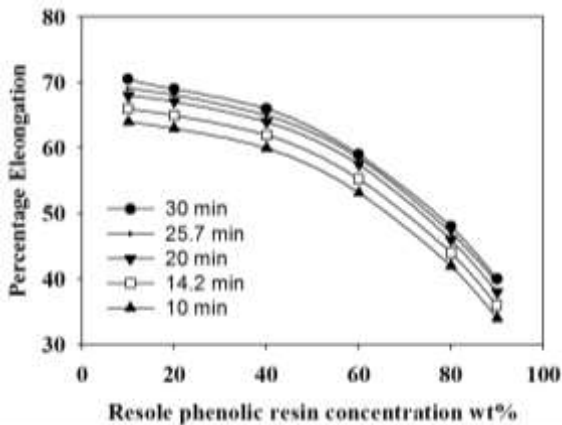


Fig. 4 Percentage elongation vs. phenolic resin concentration at different curing time from 10 to 30 min and at fixed curing temperature of 162°C

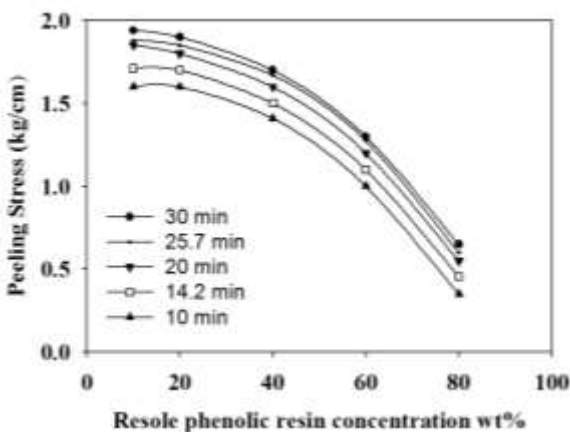


Fig. 5 Peeling stress vs. phenolic resin concentration at different curing time from 10 to 30 min and at fixed curing temperature of 162°C

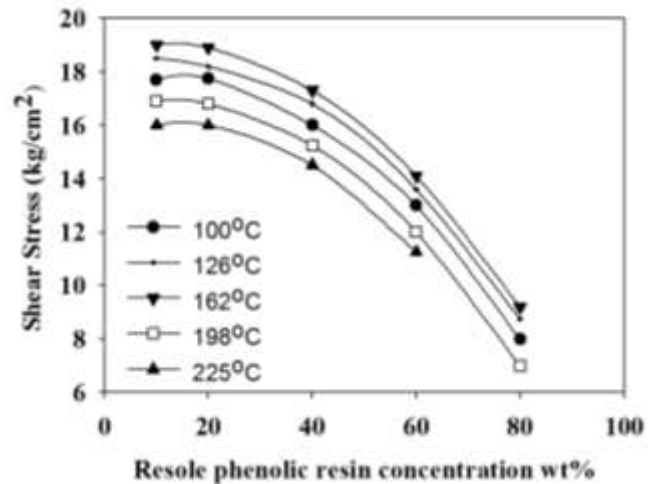


Fig. 6 Shear stress vs. phenolic resin concentration for 30 min curing at various temperatures

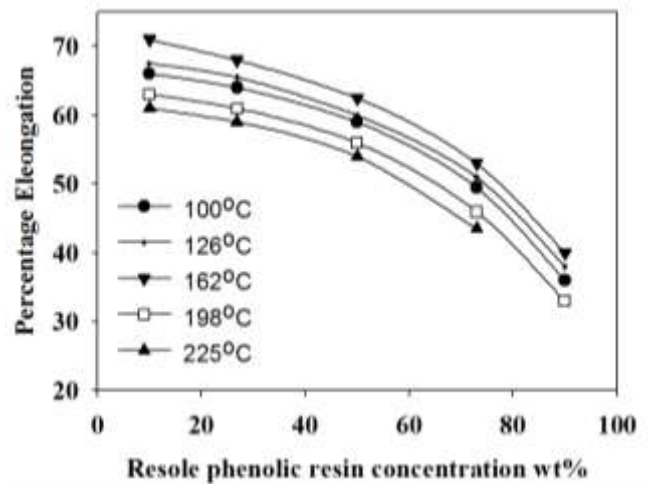


Fig. 7 Elongation vs. phenolic resin concentration for 30 min curing at various temperatures

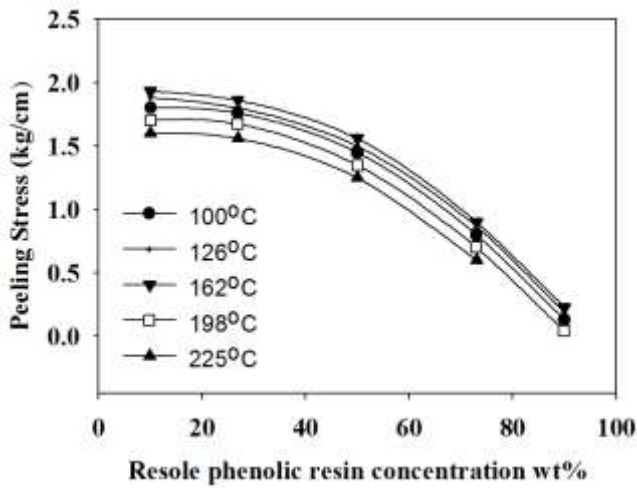


Fig. 8 Peeling stress vs. phenolic resin concentration for 30 min curing at various temperatures

From Figures 3-8, it is clear that the 10 wt% resole phenolic resin mixture gives the best mechanical properties and also increasing curing time lead to increase the crosslinking through the ether linkage between methylol group of resole phenolic resin and the hydroxyl one belongs to poly(vinyl formal) leading to an increase in the mechanical properties. the optimum concentration at curing temperature 162°C and time of 30 min increases the shear stress of poly(vinyl formal) from 15.5 to 19 kg/cm<sup>2</sup> and increase its elongation from 56% to 70% also increase the peeling stress from 1.45 kg/cm to 1.9 kg/cm, which make it has a good capability to withstand stress and possesses high elasticity and adhesion properties which renders the material suitable for coating.

D. Electrical Properties

Electrical properties investigated in this study are divided into three tests (dielectric constant  $\bar{\epsilon}$ , dissipation factor D, and loss factor  $\bar{\epsilon}$ ). In these tests a study of the effects of resole phenolic resins concentration on electrical properties after fixing curing conditions at 30 min. and 162°C has to be carried out, since it exhibits the best mechanical properties. Figures 9, 10, and 11 show the dependence of the dielectric, dissipation factor, and the loss factor on the frequency such that, increasing frequency will lead to decrease the polarity (dielectric constant) of the material due to the atomic, electronic, and directional polarization. These three types need to appear along period of time (t) and this means that it appears at a low frequency, after that these three types disappear gradually when increasing frequency and that will lead to decrease in the dielectric constant of the material. The increase of the concentration of resole phenolic resin leads to increase the dielectric constant of the mixture because of the high polarity of resole phenolic resin compared with poly(vinyl formal).

The dissipation factor was observed to increase when increasing resole phenolic resin concentration, therefore low

concentration is required to lower the energy dissipated through the insulation. The material used for electrical coating should be with consumed energy that does not exceed 0.05 [6].

Figure 11 shows how loss factor changes with respect to log F for poly(vinyl formal) mixtures 10 and 90 wt%. The loss factor  $\bar{\epsilon}$  of insulating material is commonly estimated form the following relationship [7].

$$\bar{\epsilon} = D \bar{\epsilon} \tag{1}$$

Where D is the dissipation factor and  $\bar{\epsilon}$  is the dielectric constant.

Figure 11 shows that 90 wt% poly(vinyl formal) mixture can be used in the frequency of (1000 Hz) because it gives a maximum loss at this frequency.

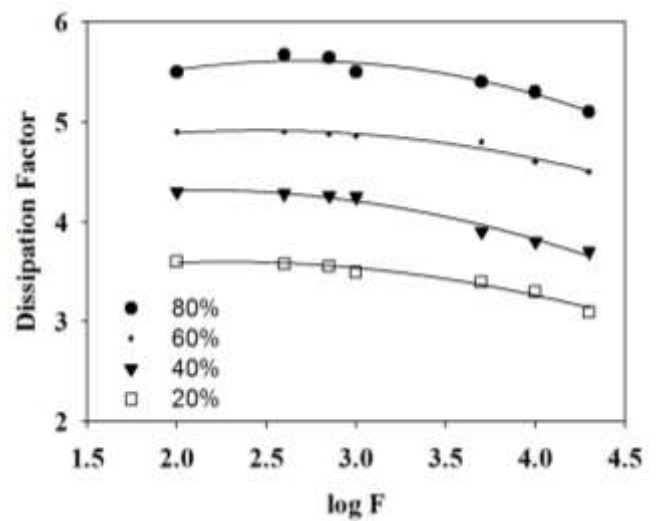


Fig. 9 Dielectric constant vs. frequency at different concentrations of resole phenolic resin and optimal conditions

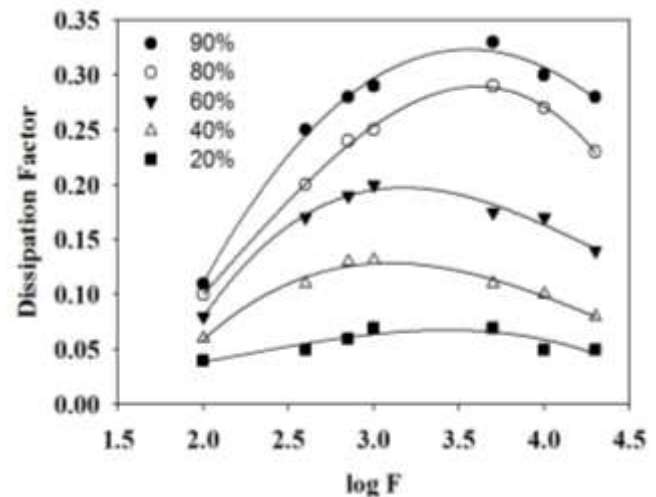


Fig. 10 Dissipation factor vs. frequency at different concentrations of resole phenolic resin and optimal conditions



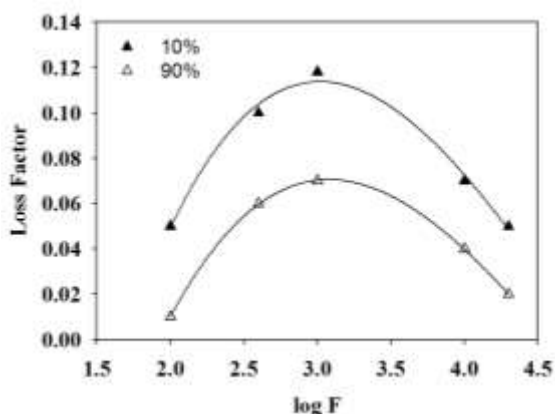


Fig. 11 Loss factor vs. frequency at different concentrations of resol phenolic resin and optimal conditions

### E. Thermal Properties

Thermal properties consist of one single test that is the Thermo-Gravimetric-Analysis (TGA) which is a dynamic technique in which the weight loss of the sample is continuously measured while its temperature is increased at a constant rate. In this test the effect of resol phenolic resin concentration on thermal properties after fixing curing conditions optimal conditions form mechanical properties, i.e., 30 min curing time and temperature of 162°C.

Figure 12 shows how weight loss changes with respect to temperature for different concentration of resol phenolic resin 0, 10 wt%. An increase in resol phenolic resin concentration leads to more thermal stability of the mixture; in other word increasing resol phenolic resin will result in a less weight loss because it consists of three dimensional cross-linked network [5] that makes it more thermally stable than poly(vinyl formal) whose thermal stability depends upon the formal content.

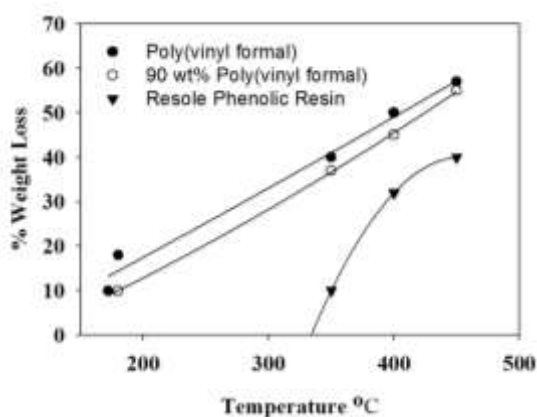


Fig. 12 Weight loss changes with respect to temperature for Poly(vinyl formal), 90 wt% poly(vinyl formal) mixture, and Resol phenolic resin

Figure 12 reveals that resol phenolic resin started to decompose at a temperature of 350°C and the weight reaches 40% at 450°C. On the other hand poly(vinyl formal) starts to decompose at 172°C and the loss is shown to be of linear increase that gives about 60% weight loss at 450°C. These

results show that resol phenolic resin has a higher thermal stability than poly(vinyl formal) and the higher the concentration of resol phenolic resin the better thermally stable mixture. however, increasing resol phenolic resin concentration will compromise the electrical and mechanical properties; therefore 10 wt% resol phenolic resin suffices to obtain an optimum mixture that balances all the mechanical, electrical and thermal properties.

### IV. CONCLUSIONS

from the present study, It is confirmed that poly(vinyl formal) can be prepared with higher formal content up to 82.3wt% from commercially available poly(vinyl acetate), acetic acid and formaldehyde by solution polymerization technique consist of hydrolysis of poly(vinyl acetate) to poly(vinyl alcohol) which is subsequently converted by formalization to poly(vinyl formal).

Acetic acid at room temperature is suitable solvent for dissolving poly(vinyl formal) to be reacted with resol phenolic resin in order to give suitable homogenous solution [3].

Resol phenolic resin is compatible with poly(vinyl formal) in order to manufacture a tough adhesive and heat resistant wire enamel.

The best conditions in which poly(vinyl formal) is compatible with resol phenolic resin is by using 10wt% of resol phenolic cured at 30 min and 162°C which give a best mechanical, electrical and thermal properties.

The prepared sample of Redux can be suitable used for wire enamel according to evaluation of most of if mechanical, electrical, physical and thermal properties which give a results better than those of the standard commercial grades of poly (vinyl formal) alone.

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