# Optimization of Halogen Free Flame Retardant Wire and Cable Compounds

**Vidya Gupta & Devendra Jain**

**Pluss Polymers Pvt. Ltd.**
**Gurgaon, Haryana, India, 122016**

## Abstract:

Halogen Free Flame Retardant [HFFR] wire and cable compounds use ATH or MDH as the main flame retardant component. Together with this it is necessary to use a maleated coupling agent and other additives such as lubricant, antioxidant, which allow incorporation of high ATH loading. Optim® maleic anhydride grafted resins can be used as coupling agents between polymers and fillers to increase the filler acceptability of polymers. Our earlier presentation (at Plexium 2007 Conference in Pune) has given a brief overview of HFFR compounds. Now we present an approach for optimization of various additives of HFFR compounds using Design of Experiments [DOE]. Statistical design approach provides an efficient framework for systematically gaining information concerning the flame retardant behavior of a polymer system. This paper describes the use of statistically designed experiments to optimize HFFR wire and cable compounds.

**Keywords**: HFFR compounds, Maleic Anhydride Grafted Polymers, Coupling Agents, Design of Experiments (DOE)

## 1. Introduction:

Polyolefins have many desirable properties like excellent electrical properties, ease of processing and adequate mechanical properties. They are used in many fields such as housing, transport or engineering applications. However, these polymers are easily flammable and therefore, imparting flame retardancy is important.

Halogen based flame retardants have been used to produce flame retardant polyolefins for many years but they give off heavy smoke and hazardous gases during combustion. In recent years, non halogen flame retardant such as Aluminum Tri Hydroxide (ATH) and Magnesium Di Hydroxide (MDH) have been considered as alternatives to halogen based flame retardants [1-4]. The use of halogen free flame retardant (HFFR) is widespread due to the increasing concern about the health and environmental risks [5].

---

**vidyagupta@pluss.co.in & djain@pluss.co.in**

The inorganic flame retardant decomposes endothermally releasing water around the temperature at which polymers themselves decompose and do not induce the smoke corrosive gas problems [6]. The drawback of inorganic flame retardants is in their high amount of loading required in polyolefins to achieve adequate flame retardancy [7]. High loading of filler leads to processing difficulties and poor mechanical properties due to incompatibility between the filler and polymers.

The usage of compatibiliser and coupling agents is one of the main approaches to solve these problems. Polymeric compatibilisers such as maleic anhydride functionalized polyolefins improve the adhesion between filler particles and matrix [8]. Reactive groups interact readily with functional groups on the inorganic filler and long hydrocarbon tails are able to anchor the polymer matrix through physical entanglements and van der Waal's interactions [9].

Statistically designed experiments can be used to optimize halogen free flame retardant (HFFR) compounds [10-12]. In this paper, we describe an approach for optimization of various additives of HFFR compounds using Design of Experiments (DOE). The One Minute DOE software was used in the statistical analysis to process the data. The statistical analysis was performed in the form of "analysis of variance" (ANOVA). This analysis includes the F-test (overall model significance) and its associated probability (p-value) for studying the most influential factor affecting the performance of the HFFR compound.

## 2. Experimental:

### 2.1 Materials

The polymers used in this study were linear low density polyethylene (LLDPE), MFI- 3.7 g/10 min; ethylene vinyl acetate copolymer (EVA), MFI-2.5 g/10 min with 18% of VA content and ethylene butene copolymer (EBC), MFI- 1.2 g/10 min. ATH particle size 0.6~3.2 microns was obtained from Nabaltec and Maleic anhydride grafted polyethylene **OPTIM® E-119** from Pluss Polymers (P) Ltd. Magnesium Stearate, Silicon gum (Genioplast), and Antioxidant (Kenox 10) were the other additives used.

### 2.2 Compositional Details

The Compositions are based on Design of Experiment (DOE), in which the number of experiments are equal to $2^n$ (two level factorial design). Where 'n' is the number of variables. Two sets of experiments were prepared to see the effect of polymers and additives. The variables in the compositions of HFFR compounds are:

**Set I**

ATH-50, Optim-5, Paraffin Oil-1, Antioxidant-0.1 [In Parts]

| | | **Low level (-)** | **Base line (0)** | **High level (+)** |
|---|---|---|---|---|
| (A) | EVA: | 10 | 15 | 20 |
| (B) | LLDPE: | 10 | 15 | 20 |
| (C) | EBC: | 10 | 17.5 | 25 |

**Table I**

| **Experiment No.** | **(A) EVA** | **(B) LLDPE** | **(C) EBC** |
|---|---|---|---|
| 1 | + | - | - |
| 2 | - | + | - |
| 3 | - | - | + |
| 4 | + | + | - |
| 5 | + | - | + |
| 6 | + | + | + |
| 7 | - | + | + |
| 8 | - | - | - |
| Base Line 1 | 0 | 0 | 0 |
| Base Line 2 | 0 | 0 | 0 |
| 1R | + | - | - |

**Set II**

ATH-60, LLDPE-10, EVA-10, EBC-14.35, Antioxidant-0.1[In Parts]

| | **Low level (-)** | **Base line (0)** | **High level (+)** |
|---|---|---|---|
| (A) Optim: | 2 | 4 | 6 |
| (B) Silicon: | 0.1 | 0.55 | 1.0 |
| (C) Mg Stearate: | 0 | 1 | 2 |

**Table II**

| **Experiment No.** | **(A) Optim** | **(B) Silicon** | **(C) Mg. Stearate** |
|---|---|---|---|
| 1 | + | - | - |
| 2 | - | + | - |
| 3 | - | - | + |
| 4 | + | + | - |
| 5 | + | - | + |
| 6 | + | + | + |
| 7 | - | + | + |
| 8 | - | - | - |
| Base Line 1 | 0 | 0 | 0 |
| Base Line 2 | 0 | 0 | 0 |
| 8R | - | - | - |

### 2.3 Preparation of Blends and Characterizations:

All formulations were compounded on a two roll mill followed by extrusion to get a good dispersion of ATH in the polymer. Then material was compression molded at 160ºC and 25 tons pressure to obtain 1 mm thick sheets of 12.5x12.5 cm.

Mechanical properties were measured using Universal Testing Machine. The test specimen size was referenced to Indian standard for cables- IS: 10810[Part 7]. The stretch rate was 250 mm/min.

Limiting Oxygen Index (LOI) of samples was measured as the oxygen concentration (%) according to ASTM D2863. Melt Flow Index (MFI) was measured at 160ºC and 21.6 kg load according to ASTM D1238. Density measured at 27ºC according to ASTM D792.

## 3. Results and Discussions

We studied the effect of various polymers on the mechanical properties of the HFFR compounds in Set I. The mechanical properties, MFI and densities are represented in Table III. Total inorganic filler content was set to 50 parts. The tensile properties of Set I of the HFFR compounds was lower due to the poor interfacial properties between the filler and the matrix. The elongation properties of the HFFR compounds for Set I was higher when EBC was present at a higher level. This is due to the elastomeric nature of the ethylene butene copolymer.

**Table III**

| Ex. No. | EVA | LLDPE | EBC | T.S. | Elon. | MFI | Density | LOI |
|---|---|---|---|---|---|---|---|---|
| 1 | 20.00 | 10.00 | 10.00 | 95.00 | 66.00 | 13.31 | 1.40 | |
| 2 | 10.00 | 10.00 | 10.00 | 107.00 | 44.00 | 9.14 | 1.24 | |
| 3 | 10.00 | 20.00 | 25.00 | 86.00 | 46.00 | 9.90 | 1.25 | |
| 4 | 20.00 | 20.00 | 10.00 | 92.00 | 105.00 | 12.00 | 1.26 | |
| 5 | 20.00 | 20.00 | 25.00 | 82.00 | 223.00 | 12.18 | 1.15 | |
| 6 | 10.00 | 10.00 | 25.00 | 85.00 | 135.00 | 10.47 | 1.29 | 24.6 |
| 7 | 10.00 | 20.00 | 10.00 | 97.00 | 53.00 | 8.29 | 1.21 | |
| 8 | 20.00 | 10.00 | 25.00 | 104.00 | 514.00 | 10.62 | 1.38 | |
| 9 | 20.00 | 10.00 | 10.00 | 84.00 | 70.00 | 9.80 | 1.10 | |
| BL1 | 15.00 | 15.00 | 17.50 | 92.00 | 67.00 | 12.00 | 1.34 | |
| BL2 | 15.00 | 15.00 | 17.50 | 86.00 | 127.00 | 10.93 | 1.44 | |

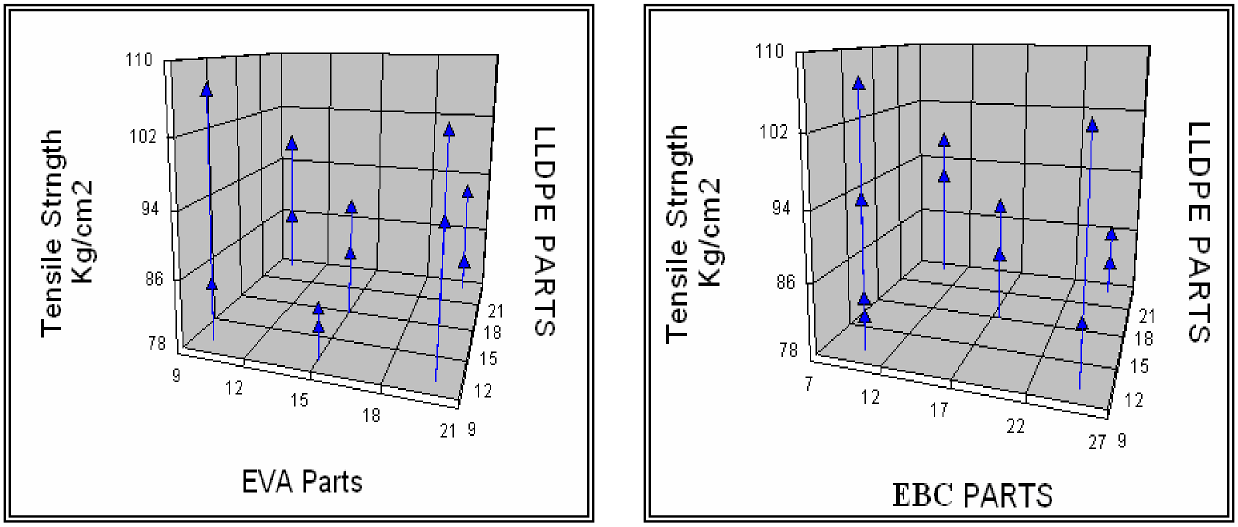


**Fig. 1 Effect of Polymers in Tensile Strength**

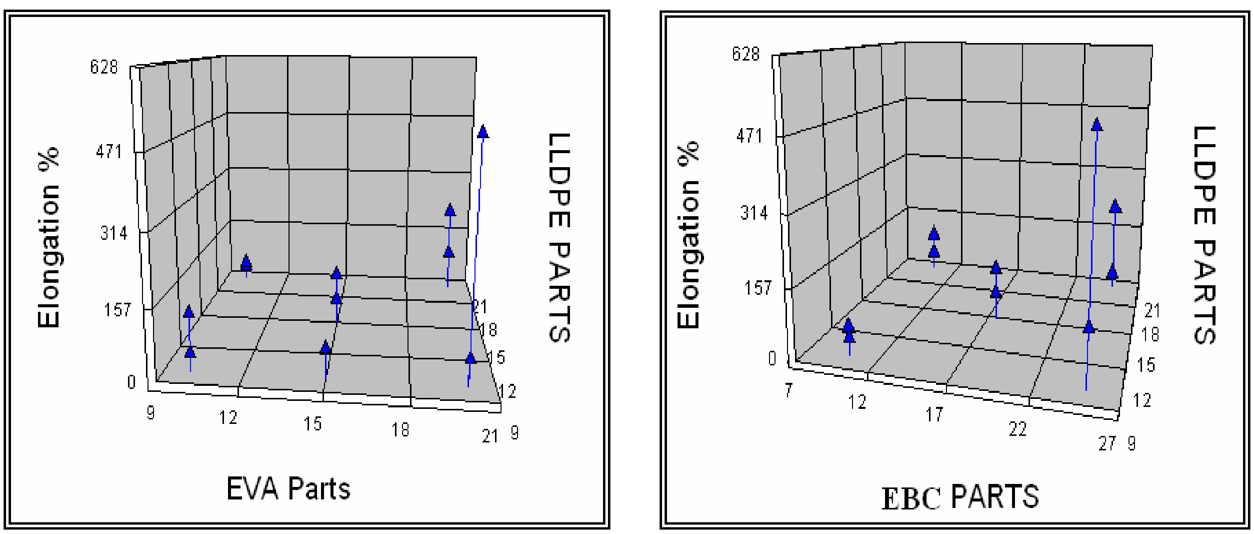


**Fig. 2 Effect of Polymers in Elongation**

Figures 1 and 2 show the effect of polymers in tensile strength and elongation. The statistical analysis shows that the p-value is 1.00 for tensile strength, which is higher than the significance level ($\alpha = 0.05$) meaning that the various ratios of the polymer has no effect on tensile strength of the HFFR compound. However, for elongation the p-value is 0.0449 for EVA and 0.0304 for EBC which indicates the ratios of EVA and EBC have significant effect on the elongation properties of the HFFR compound. An increase in the EVA and EBC parts in LLDPE increases the elongation of HFFR compounds.

The LOI value of the compound indicates that the 50 parts of ATH was insufficient to make these halogen free compounds flame retardant.

Table IV represents the mechanical properties, MFI and densities of Set II compounds. We studied the effect of coupling agent OPTIM® and other additives on the mechanical properties of the HFFR compounds. The total filler loading was 60 parts to obtain LOI values desired of the HFFR compounds. The coupling agent was used to improve the bonding of filler to polymer. OPTIM® maleic anhydride grafted resins may be used as coupling agents between polymers and fillers to increase the filler acceptability of polymers. **OPTIM® E-119** from Pluss Polymers provides excellent coupling action between the filler and the polymer matrix. The tensile strength and elongation was highest when Optim® coupling agent is at higher level and silicon at lower level, which is contradictory. It is proposed to investigate this further. Our aim in this work is to be able to use DOE for designing a commercially attractive HFFR compound and subsequently in other compounding applications.

**Table IV**

| Ex. No. | OPTIM | Silicon | Mg Stearate | Tensile | Elongation | MFI | Density | LOI |
|---|---|---|---|---|---|---|---|---|
| 1 | 6.00 | 0.10 | 0.00 | 89.00 | 111.00 | 4.66 | 1.49 | |
| 2 | 2.00 | 1.00 | 0.00 | 80.04 | 130.00 | 4.51 | 1.45 | |
| 3 | 2.00 | 0.10 | 2.00 | 65.00 | 150.00 | 5.1 | 1.36 | |
| 4 | 6.00 | 1.00 | 0.00 | 108.00 | 152.50 | 4.8 | 1.49 | |
| 5 | 6.00 | 0.10 | 2.00 | 82.20 | 130.00 | 3.8 | 1.45 | 27.7 |
| 6 | 6.00 | 1.00 | 2.00 | 80.00 | 142.00 | 4.5 | 1.41 | |
| 7 | 2.00 | 1.00 | 2.00 | 64.10 | 67.00 | 3.6 | 1.44 | |
| 8 | 2.00 | 0.10 | 0.00 | 111.50 | 162.50 | 3.9 | 1.46 | |
| BL1 | 4.00 | 0.55 | 1.00 | 65.56 | 150.00 | 3.6 | 1.41 | 29.4 |
| BL2 | 4.00 | 0.55 | 1.00 | 67.00 | 150.00 | 5.5 | 1.46 | |
| 8R | 2.00 | 0.10 | 0.00 | 108.00 | 270.00 | 3.8 | 1.34 | |

Figures 3 and 4 show the effect of coupling agent OPTIM® and other additives on the tensile strength and elongation of the HFFR compounds. The statistical analysis showed that the p-value was 0.0373 for tensile strength, which is lower than the significance level ($\alpha = 0.05$) meaning that the ratios of Mg Stearate has significant effect on the tensile strength of the HFFR compound. The tensile properties decrease when Mg Stearate is in higher level with OPTIM® coupling agent and silicone gum. The p-value for elongation properties was 1.00 which is higher than the significance level ($\alpha = 0.05$) meaning that the various ratios of the additives have no effect on elongation of the HFFR compound.

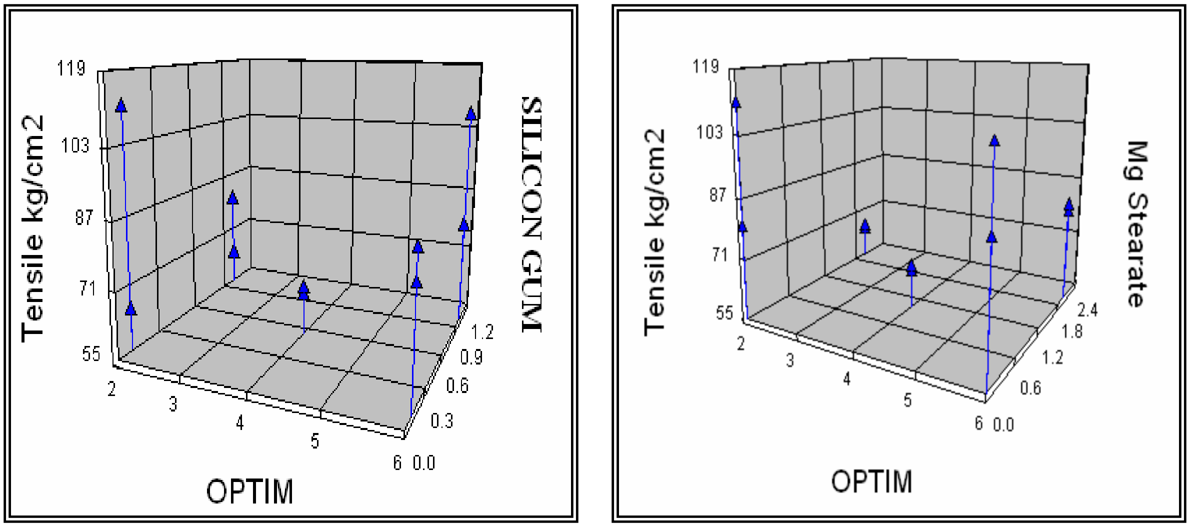


**Fig. 3 Effect of coupling agent Optim® and additives in Tensile Strength**

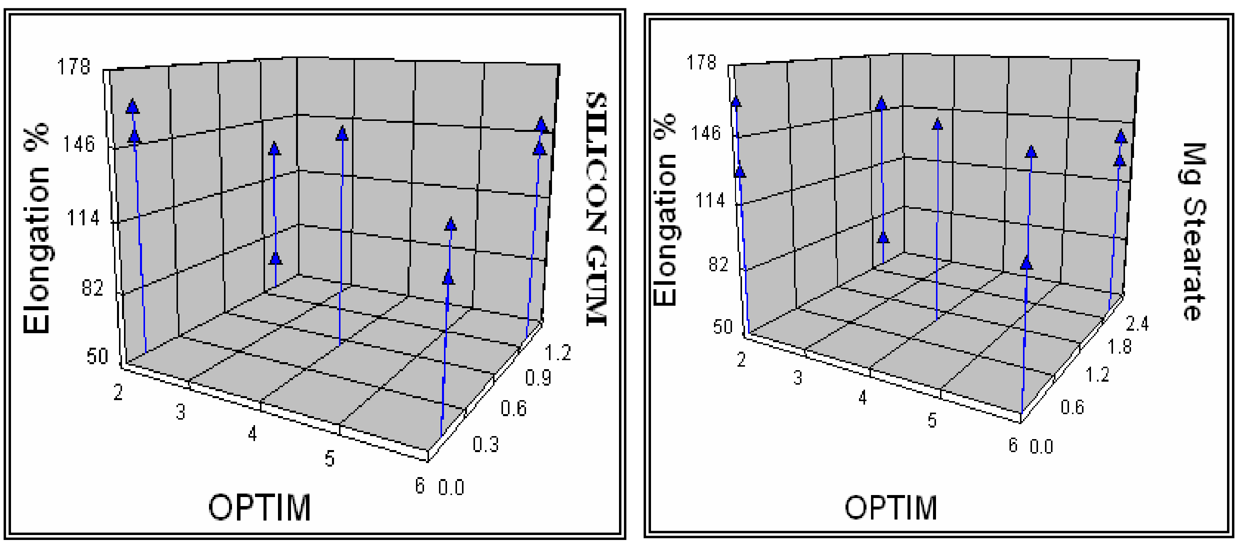


**Fig. 4 Effect of coupling agent Optim® and additives in Elongation**

The LOI value of the compound indicates that a minimum 60 parts of ATH is needed to make flame retardant HFFR compounds.

## 4. Summary

In this study, we described an approach for optimization of composition of HFFR compounds using design of experiments (DOE). The mechanical properties, MFI and densities were measured with various polymers. Also the effect of coupling agent OPTIM® and other additives on the tensile strength and elongation of the HFFR compounds was studied. We also believe that two roll mill used does not give adequate mixing and hence Set II is proposed to be validated by use of an internal mixer. Our further work will be focused on the effect of various OPTIM® coupling agents, other processing aids and various polymer compositions to get a commercially attractive HFFR compound.


## Acknowledgement

We gratefully acknowledge the helpful discussions and assistance received by Mr. Neeraj Jain of Matrix Polytech (P) Ltd., Delhi during the course of this study.


## References


1. G.Wang, P.Jiang and Z.Zhu; Polymer Composites; 23; 691; 2002.
2. J.T.Yeh, M.J.Yang and S.H.Hsieh; Polym Degrd Stb; 61; 465; 1998.
3. A. Szep A, A. Szabo , N Toth , P Anna, G Marosi; Polym Degrd Stb;91; 593; 2006.
4. G. Beyer; Fire Mater; 25; 193; 2006.
5. IH.Kim, GJ.Nam and GJ. Lee; Antec; 1819; 2006.
6. LS Birnbaum, DF. Staskal; Environ Health Perspect; 112; 9; 2004.
7. L. Haurie, AI.Fernandez, JI. Velasco, JM Chimenos, JML Cuesta and F.Espiell; Polym Degrd Stb.; 92; 1082; 2007.
8. K. Hausmann, V.Flaris; Polym and Polym Comp; 5; 113; 1997.
9. F.Rahma, S.Fellahi; Polym Comp; 21; 175; 2000.
10. ED.Weil, W.Zhu, N.Patel & SM. Mukhopadhyay; Polym Degrd Stb; 54; 125; 1996.
11. LM.Babcock, M.Altekar; Chemom. and Intell. Labo. Sys; 29; 141; 1995.
12. H.Dvir, M.Gottlieb, S.Daren, E.Tartakovsky; Compo Sci and Tech; 63; 1865; 2003.