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OPTIMIZATION OF MANUFACTURING PROCESSES AND WORK ENVIRONMENT

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CHAPTER 12

FLAME RETARDANTS FOR PLASTICS

Based on scientific and technical papers, the products and development concerning flame retardants for plastics are described. The tests of plastics on resistivity to flame ignition are specified. Three types of flame retardants: reactive, additive and intumescent are presented and discussed in details. The risks related to their use and the latest technical developments in flame retardants are also presented.

Key words: flame retardants, reactive additives, intumescent additives, flame tests, plastics

1. INTRODUCTION

Because most plastics are composed of hydrocarbons and their derivatives, they are combustible and this property creates the potential danger of fire. On the other hand, plastics are often used in aplications where safety is important, such as in transport, building, appliences and also in electrical and electronic equipment and in these conditions the materials should be nearly incombustible or at least very difficult to ignite and to burn.

Underwriters Laboratories, a leading organization of product safety certification, created UL 94 standard to test and classify flame safety of materials used for appliences and electronic equipment. There are 12 flame classifications specified in UL 94 that are assigned to materials based on the results of these small-scale flame tests. These classifications listed in descending order for each of the following three groupings are used to distinguish a material's burning characteristics after test specimens have been exposed to a specified test flame under controlled laboratory conditions.

Six of the classifications relate to materials commonly used in manufacturing enclosures, structural parts and insulators found in consumer electronic products (5VA, 5VB, V-0, V-1, V-2, HB). Three of the remaining six classifications relate to low-density foam materials commonly used in fabricating speaker grills and sound-deadening material (HF-1, HF-2, HBF). The last three classifications are assigned to very thin films, generally not capable of supporting themselves in a horizontal position (VTM-0, VTM-1, VTM-2). These are usually assigned to substrates on flexible printed circuit boards.

Specimens molded from the plastic material are oriented in either a horizontal or vertical position depending on the specifications of the relevant test method and they are subjected to a defined flame ignition source for a specified period of time. In some tests, the test flame is only applied once as is the case of the horizontal burning (HB) test, while in other tests the flame is applied at least twice. An HB flame rating indicates that the material was tested in a horizontal position and found to burn at a rate less than a specified maximum.

The three vertical ratings, V2, V1 and V0 indicate that the material was tested in a vertical position and self-extinguished within a specified time after the ignition source was removed. The vertical ratings also indicate whether the test specimen dripped flaming particles that ignited a cotton indicator located below the sample. UL 94 also describes a method in which the test flame is applied for up to five applications in testing for a 5VA or 5VB classification. These small-scale tests measure the propensity of a material to extinguish or spread flames once it becomes ignited.

When looking at the flame ratings for plastic materials commonly molded to fabricate enclosures, structural parts and insulators found in consumer electronic products (5VA, 5VB, V-0, V-1, V-2 and HB), a material classified as 5VA or 5VB is subjected to a flame ignition source that is approximately five times more severe than that used in the V-0, V-1, V-2 and HB tests. Also, the specimens may not drip any flaming particles. The three remaining six classifications specified in UL 94 relate to low-density foam materials commonly used in fabricating speaker grills and sound-deadening material (HF-1, HF-2, HBF). The remaining three classifications are assigned to very thin films commonly used in flexible printed wiring boards, generally not capable of supporting themselves in a horizontal position (VTM-0, VTM-1, VTM-2). A flame rating of VTM-0 cannot be considered equivalent to a V-0 rating as the test methods are quite different. Likewise, VTM-1 and VTM-2 cannot be considered equivalent to V-1 and V-2 respectively.

In order to decrease the danger of fire of plastics, polymers are supplemented with materials that inhibit or resist fire and even though some polymers like polyvinyl chloride are themselves to some extent resistant to burning, even they must be additionally supplemented with these additives, called usually flame retardants (FR) or flame suppression additives. These additives may work by quenching a flame by depriving it of oxygen or they may absorb heat and produce water and in that way reduce the temperature.

Flame retardant additives are used in various combinations, often exhibiting a synergistic effect.

Experience has shown that fire itself is not the gretest danger; more dangerous to people are the toxic by-products formed during combustion and dense smoke that creates problems for people to escape in time. The control of smoke and toxic products during combustion of plastics is becoming important in assessing FR additives.

Flame retardant additives may be divided into three types:

1. Reactive flame retardants, which are usually added during the polymerization stage and are co-polymerized with monomers;
2. Additive flame retardants, which are added after polymerization stage during a subsequent compounding stage;
3. Intumescent additives, which under high heat swell and form an insulation protecting the plastic.

2. REACTIVE FLAME RETARDANTS

Reactive FR are mostly used in thermosetting resins, such as unsaturated polyesters and epoxide resins. For polyesters, the main reactive retardants are chlorendic acid (HET acid) and dibromoneopentyl glycol (DBNPG).

Chlorendic acid (HET acid) is a white crystalline material with chemical formula $C_9H_4Cl_6O_4$. It is also called Hetron 92, Hetron 92C or hexachloro-*endo*-methylenetetrahydronaphthalic acid. This acid is also used as a finishing flame-retardant treatment for wool. A major use of HET is in production of fiberglass-reinforced resins for chemical industry equipment. When reacted with nonhalogenated glycols, it forms halogenated polyols used as flame retardants in polyurethane foams. Chlorendic acid is also used for production of dibutyl chlorendate and dimethyl chlorendate, which are also used as reactive flame retardants in plastics. When chlorendic acid is used in polymers, whether as a curing agent or as a flame retardant, it bonds covalently to the polymer matrix, which reduces its leaching to the environment. In Europe, 80% of chlorendic acid is used in production of flame-resistant composites for building and transportation. In the USA, Latin America and Asia, 20-30% is used in flame retardant applications and the rest is used in corrosion-resistant plastics.

Dibromoneopentyl glycol (DBNPG) is a reactive flame retardant containing 60% aliphatic bromine. Thermosetting polyester resins can be formulated with this over a wide range of compositions to provide a broader selection of resin properties than are available with anhydride flame retardants. Resins formulated with types of DBNPG have high chemical and flame resistance, minimal thermal discolouration and excellent light stability. It can also be used with polyurethane rigid foams.

Tribromoneopentyl alcohol (TBNPA) is a reactive flame retardant containing more than 70% aliphatic bromine. It is exceptionally stable and is particularly suitable where thermal, hydrolytic and light stability are required. It is highly soluble in polyether potyols, making it particularly suitable for use in polyurethane polymers.

Tetrabromobisphenol A (TBBPA) $C_{15}H_{12}Br_4O_2$ is produced mainly in the USA, Japan and Israel; together about 150 000 tonnes per year. The main trade marks are Saytex 111, Saytex RB-100, Fire Guard 2000, Firemaster BPA. It is mostly used in epoxy resins and polycarbonates. Albemarle is constructing a worldscale (50 000 tonnes/year) plant to produce the next generation of Saytex CP- 2000, using continuous process technology that will "virtually eliminate" variations arising from small-batch production.

Brominated flame retardants are supposed to be more efficient than chlorendic acid (which has also become more expensive). With epoxies, the best system (based on present evidence) appears to be reactive phosphorus organic compounds, which are toxicologically harmless in fire and are chemically linked to the resin matrix, so that mechanical and chemical properties of plastic are not affected.

3. ADDITIVE FLAME RETARDANTS

Additive flame retardants are more frequently used than reactive FR. The use of particular FR depend on the precise conditions in which the additive is expected to operate and also on its price. The main additive flame retardants are inorganics (aluminium trihydrate, antimony trioxide, magnesium hydroxide, phosphorous compounds), halogenated organic compounds and some nitrogen compounds.

Aluminium trihydroxide $Al(OH)_3$, called also aluminium trihydrate (ATH) or hydrated alumina is the most widely used flame retardant, representing 43% of all flame retardant chemicals in quantity (but about 29% in value, it is relatively inexpensive). In addition to flame retarding and smoke suppressing properties, it is also an economical filler. This compound begins to decompose at temperatures above 180°C, with an endothermic reaction that absorbs energy; this has the effect of decreasing the rate of heat release from a burning polymer filled with aluminium trihydroxide. At high temperatures it also releases water vapour, which dilutes the combustion gases and toxic fumes. It is also an excellent smoke suppressant, partly due to the fact that a high loading naturally reduces the amount of available combustible material, but also because the high surface area aluminium oxide formed during combustion will also adsorb fine smoke particles, and will act to catalyze cross-linking reactions, promoting formation of a solid char rather than smoke. It is used mainly in unsaturated polyesters in the building (construction) industry and in cable sheathing compounds. Its use is limited by a maximum processing temperature of about 200°C. The high content of aluminum trihydrate in plastic needed to achieve good flame retading performance can be detrimental to mechanical and electrical properties. It is not stable at high temperature due to loss of water. To improve the performance, surface-modified grades have been developed with enhanced processability and chemical coupling.. Selected surface modifications (particularly based on organo-functional silanes) can also improve specific properties, by increasing the ATH/polymer interracial adhesion. Typical improvements are flame/smoke properties, mechanical properties (including increased tensile, flexural, impact and elongation) and better resistance to water permeation, which may improve electrical properties. Controlled viscosity and ultra-low viscosity grades (allowing exceptionally high loadings) are among new developments (by Alcan), while modification of particle shape with a reduction in coarse particles offers many advantages during compounding. Surface-modified grades by Huber Engineered Minerals (Hymod) give better dispersion with increased compatibility with the resin matrix, resulting in lower viscosity or increased loading, for improved processing and properties. Another development is a wetting/dispersing and low profile additive by BASE which made it possible to formulate a polyester SMC with ATH to meet the stringent French Railway M 1/F0 fire rating, without exceeding the recommended viscosity level.

Solid magnesium hydroxide has also smoke suppressing and fire retarding properties. This is due to the endothermic decomposition it undergoes at 332°C $Mg(OH)_2 \rightarrow MgO + H_2O$.

The heat absorbed by the reaction acts as a retardant by delaying ignition of the associated substance. The water released dilutes any combustible gases and inhibits oxygen from aiding the combustion. Another mineral that is used in similar fire retardant applications is hydromagnesite.

Magnesium hydroxide can impart flame retardance and smoke suppression to a wide variety of thermoplastics and elastomeric formulations. It is temperature stable to 330°C, allowing processing with a wide variety of thermoplastics and use where aluminium trihydrate is not sufficiently stable. Lower smoke ratings than with halogenated additives can usually be obtained. It is used particularly in cable sheathing, polypropylene and polyamides.

Prospects for the use of magnesium hydroxide are generally good. Magnesium hydroxide may replace aluminium-based products, especially if aluminium prices rise. Magnesium hydroxide is attracting interest, with current consumption of 3175 tonnes forecast to rise to 4080 tonnes in the next five years.

Antimony trioxide is often used because it has a synergistic effect with most halogenated flame retardants. It is also used in plasticized PVC because of its synergy with chlorine. Antimony oxide should not be used if translucency is required. In some cases ferric oxide is used in its place for similar physical properties but improved electrical properties. Antimony trioxide has been shown by extensive research to be non-carcinogenic. However, after years of low prices, antimony trioxide reached a high price of $8.80/kg in 1996-7 and later leveled out at about $6 per kg and the growth of its use has slackened considerably.

The annual consumption of antimony trioxide in the United States and Europe is approximately 10 000 and 25 000 tonnes, respectively. The main application is for flame retardants in combination with halogenated materials. The combination of the halides and the antimony being key to the frame-retardant action for polymers helps to form less flammable chars. Such synergisticflame retardants are found in electrical apparatus, textiles, leather and coatings.

Phosphorus flame retardants are also used in the US market to complement standard halogenated products. Further development of halogen/phosphorus synergism could lead to wider use as a substitute for antimony oxide.

Halogenated flame retardants are mainly chlorinated or brominated compounds. Economically they are the most important, but due to pressure from environmental activists the use of chlorinated retardants has been sharply reduced and the attack has now turned to brominated FR compounds.

Brominated FR more numerous than chlorinated, because their efficiency is significantly better due to the fact that bromine is heavier than chlorine and decomposition products are less volatile at high temperature. The plastics industry claims that there is no evidence to support the belief that dangerous compounds are released during their incineration and the safety of brominated retardants has been confirmed by institutions such as the US Environmental Protection Agency, the World Health Organization and the Organization for Economic Cooperation and Development. Nevertheless, some brominated compounds have been withdrawn from production. Grades have been developed for use with high impact polystyrene, polyolefins and engineering plastics. Tests on a typical grade (Saytex 8010, from Albemarle) showed no detectable quantities of brominated dioxins or furans and thus complied with the German Dioxin Ordinance. Polystyrene resins using a blend of brominated FR and antimony trioxide can be safely recycled.

In the USA bromine compounds have the strongest growth of all FRs and are the second largest FR additive group, while the world bromine industry forecasts a steady 8% per year growth, largely driven by increased use in plastics (which make up 30% of consumption). In Europe they continue to dominate the electronic circuit board sector making them the most valuable sector of the industry at present, accounting for around 34% of total sales by value.

New brominated FRs are being introduced almost daily, making it impossible to give a comprehensive list. The company Great Lakes, the leading manufacturer of brominated FR, lists 26 grades in six main chemical types. Other manufacturers are working on other types. These are the main groups:

a) polybrominated diphenyl oxide (PBDO or PBDPO) or polybrominated diphenyl ethers (PBDE) having the formula $C_{12}H_{10-x}OBr_x$, which are suitable for most plastics, except polystyrene foam. The most important compounds are penta-DBE, octa-BBE and deca-DBE. They have an uncertain future because of fears about possible air pollution during the incineration of plastics waste.

b) dibromostyrene and derivatives: includes graft copolymers with polypropylene; they are recommended with ABS and styrenes, most engineering thermoplastics, unsaturated polyester resins and polyurethane foams, but they are not recommended for PVC, PS foam and rigid PU foam.

c) Hexabromocyclododecane (HBCD or HBCDD) $C_{12}H_{18}Br_6$. Its primary application is in extruded polystyrene foam (XPS) and expanded polystyrene foam (EPS) that is used as thermal insulation in the building industry. HBCD is highly efficient in this application, so that very low levels are required to reach the desired flame retardancy. Typical HBCD levels in EPS are 0.7% and in XPS 2.5%. At present, according to BSEF, the brominated flame retardant industry panel, HBCD is the only suitable flame retardant for these applications and any other flame retardant would likely need higher load levels in the polystyrene foam. Other uses of HBCD are upholstered furniture, automobile interior textiles, car cushions and insulation blocks in trucks, packaging material, video cassette recorder housing and electric and electronic equipment. The industrial demand in Europe is estimated on 9500 tons per year.

d) Pentabromobenzyl acrylate (developed for engineering thermoplastics and now in full production by Dead Sea Bromine Group) can be polymerized or copolymerized in the extruder, giving UL 94 V-O ratings without loss of physical or mechanical properties in host resins such as nylon 6 and 66, PBT and polycarbonate. This polymeric additive is also available from LCL-IP as FR-1025. It is more expensive than other polybrominated aromatics, but has good electrical properties and good compatibility with fiber reinforcement.

e) Tetrabromophthallic anhydride $C_8Br_4O_2$ and derivatives are used mainly with thermosetting resins and PUs; also PVC and thermoplastic elastomers.

f) Tribromophenol and derivatives: used with ABS and styrenes, polycarbonate, polyamide, PS and PU foams and thermosetting resins; not suitable with polyolefins and PVC.

4. INTUMESCENT FLAME RETARDANTS

Intumescent materials swell to many times their original thickness at high temperature, so producing a thick insulating layer with good resistance to erosion by fire and hot gases.This fire resistant insulating foam serves to isolate heat and oxygen, extinguishing the fire.

Phosphorus compounds are often used as components of intumescent flame retardants. On heating thay form polyol phosphates, which can break down to form char. Typical formulation in coating contains a phosphoruous compound as ammonium polyphosphate, a char forming polyol like pentaerythriol, a blowing agent such as melamine and a binder. For a mixture to form an efficient intumescent system, three components are required: an inorganic acid as dehydrating agent, a carbon-rich material as char former and a blowing agent, helping to form the foam. The optimum compositions and optimum ratio for a burning material (polymer) must be determined experimentally. Nitrogen based compounds are widely used in almost all char forming flame retardant systems, because they produce less smoke and toxic gases.

Some low-toxicity alternatives to antimony trioxide in halogenated polymer systems work synergistically to form a char in conjunction with halogenated polymers. During combustion the vapour phase changes the flame chemistry to inhibit fire growth by removing free radicals which support combustion. Additional effects in the condensed phase produce advantages not seen with traditional antimony trioxide systems. A hard carbonaceous char is formed which further retards flame propagation and reduces the amount of smoke and carbon monoxide during combustion. Grades are thermally stable up to 200°C, suitable for brominated polyesters, PVC and halogenated polyethylene, or thermally stable in all polymer systems.

Zinc borate, thermally stable up to 290°C, functions mainly in the condensed phase, promoting the formation of a char, which can be enhanced by the finer particle size.

Nanocomposites are the subject of intense research for a number of properties such as improved barriers to gas, higher mechanical strength and improved flame-retardancy. Plate-like particles of special clays, one nanometre (one billionth of a metre) thick by 1000 nanometres in diameter, are being studied as flame retardants in plastics by the US National Institute for Standards and Technology (NIST), Gaithersburg, Maryland. Initial research showed that the addition of as little as 5% of nanosized clay particles could produce a 63% reduction in the flammability of nylon 6.

More recent studies have shown that flame retardancy in many other polymers can be boosted by dispersing clay at the molecular level. Other new intumescent systems include the use of expanded graphite flakes.
Recently an intumescent flame retardant additive based on melamine phosphate (Melapur 200) has been launched by DSM, and is claimed to allow production of white or coloured non-halogenated glass fibre reinforced PA66 compounds. It is said to be thermally stable at processing temperatures up to 320°C, while the compounds exhibit an E-modulus of more than 10 000 kPa, elongation at break higher than 2.1% and a Charpy impact strength of more than 40 kJ/m^2. It can be used with, or as an alternative to the nitrogen/phosphate compounds currently in use.

5. LATEST PRODUCTS AND DEVELOPMENTS

The European situation regarding polybrominated biphenyls and biphenyl oxides (PBBs and PBBOs) has been set out by the European Brominated Flame Retardant Industry Panel (EBFRIP), a Sector Group of the European Chemical Federation (CEFIC).
The Dutch Ministry of the Environment (VROM) in particular has long been concerned about PBBs and PBBOs and has considered introducing its own legislation, strongly opposed by the Dutch Plastics Federation (NFK) and against evidence by the Dutch National Institute for the Environment (RIVM) that studies of municipal waste incineration did not show any relation between bromine content of disposed waste and formation of brominated dioxins.
In 1990, the EC Commission carried out a risk analysis that concluded that, although there was a potential for release of furan from incineration of waste, this was "not confirmed by actual emission measurements". Subsequently, with the support of some countries, a proposal was published to ban use of PBBO, but the Commission later decided to halt a proposed Directive until a new proposal on improved fire safety standards in furniture and furnishings was published. At an EC meeting in 1993, all member states (except The: Netherlands) expressed their preference for a common EC policy and the Dutch plastics bodies were successful in convincing the Environment Ministry that the original risk assessment should be re-evaluated, with participation from industry. There has also been growing anxiety in Holland about the economic consequences of the proposed legislation, and it is difficult to predict the timetable - or whether legislation will be implemented at all.
Although bromines may eventually gain a clean bill of health, many manufacturers of plastics are playing it safe, and development of effective non-halogenated FR grades is a top priority; while in Japan the Ministry of International Trade and Industry (MITI) has put up a budget of about ¥20 million

in 1999 (US$ 170 000) towards development of non-halogenated FR systems, believing that the lack of them will hinder Japanese trade with the rest of the world.

In Europe, BASF is looking towards nitrogen organic compounds and magnesium hydroxide for its recently-launched Ultramid KR 4205 and 4455 polyamide, and its latest grade (KR4480) is a reinforced PA6 using a nitrogen compound, giving V-2 rating and resistant to glow wire at 960°C, with greater toughness, good stiffness and much better flow properties, at a lower density. The company is also working on PBTs: it has a non-halogenated grade (Ultradur B4000) which achieves V-2 and 960°C glow wire, with low flue gas density and high tracking resistance, and predicts that it will have a V-0 rated PBT on the market within two years.

In Japan, Teijin has launched a halogen-free polybutylene terephthalate (PBT) meeting the UL94 specification at a V-0 classification, using a phosphorus-based system and replacing antimony oxide with a special auxiliary. Kyowa Chemical Industry Co and Taheto Chemical Industries Co have also been developing non-halogenated retardants, using magnesium hydroxide.

Another new development is a series of brominated FRs, based on three reactive brominated flame retardant monomers, introduced as innovative molecules that can be used as building blocks or tailored during synthesis or compounding to modify commercial resins. They are readily soluble in styrene monomer, allowing reaction into systems such as unsaturated polyesters and can also be incorporated by reactive extrusion (for example, in a glass-reinforced PBT compound).

The use of pure fine particle talcs in combination with a bromohalogenated compound is another interesting line of research, aimed at optimizing the mechanical properties and flame resistance of polypropylene compounds. Work in France (by Ecole des Mines d'Als and Talc de Luzenac) suggests that this approach can also produce compounds with reduced corrosive products, in comparison with traditional (and more costly) solutions.

The key trends in the market are identified in a recent report on the US market (by Business Communications Co Inc). This indicates that the largest single flame retardant will continue to be alumina trihydrate, with a moderate growth rate of 3.1% per year, but continuing to offer the most cost-effective system, however, the strongest growth will be with brominated compounds, with a rate of 8.5% against a growth rate of 5% per year for the whole sector, taking them to second place. The bromines shouldl eventually overcome environmental concerns because of their unique properties and the lack of acceptable alternatives.

The interest in reducing smoke and corrosion favours the use of phosphorus-based FR compounds, which (with a growth rate of 7.0%) will become the third-largest grooup of FR additives.

Other flame retardants (mainly boron-, molybdenum- and nitrogen based compounds) will continue to find markets as synergists and partial replacements for higher priced chemicals, with increasing consumption.
There is some agreement among research agencies about the world market for flame retardants. Flame retardants are estimated to make up 31% of the world volume of performance additives (nearly 850 000 tonnes).

Technical developments in flame retardants continue as plastics components require more demanding processing and end-use performance. In electronics, the use of lead-free solder and smaller and thinner components cause an increasing exposure to high temperatures, which require the use of higher temperature polymers. The flame retardants used in these polymers must then withstand higher processing temperatures and have higher flow properties. Great Lakes' new Firemaster® CP-44HF polybrominated styrene co-polymer offers increased thermal stability and a lower molecular weight for higher flow. Albemarle's new Saytex® HP3010 flame retardant, a brominated polystyrene, offers high flow, improved colour and improved temperature stability. In automotive wire and cable applications, increasing under-the-hood temperatures are resulting in replacement of PVC with polyolefins.
The polyolefin replacements need greater flame retardancy, which is met with ATH and magnesium hydroxide FRs. Coated grades of ATH and magnesium hydroxide are being developed to improve the FR's compatibility with the resin, driven primarily by demand in automotive wire and cable applications. In some applications the coating can contain up to 60% inorganic flame retardants. Improving compatibility improves final properties of the cable, for example its flexibility.
UV stability of plastics is an increasing concern with the trend toward grey electronic enclosures. Flame retardants with improved UV stability include Albemarle's Saytex 8010, Great Lakes' Firemaster 2100, and DSBG's FR-245.
Ciba has recently launched Tinuvin® FR products which offer combined flame retardancy and light stability for a wide variety of demanding outdoor applications of polyolefins such as cladding, roofing and stadium seats.

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ŚRODKI OBNIŻAJĄCE PALNOŚĆ TWORZYW STUCZNYCH

Na podstawie przeglądu literatury naukowo-technicznej opisano stosowane produkty i najnowsze prace dotyczące środków obniżających palność tworzyw sztucznych (tzw. antypirenów). Przedstawiono podstawowe testy palności tworzyw sztucznych. Szeroko opisano zasadnicze grupy antypirenów, a mianowicie dodatki reaktywne dodawane w trakcie syntez polimerów, dodatki dodawane w trakcie procesu komponowania składu tworzywa po procesie polimeryzacji oraz dodatki obniżające palność tworzyw poprzez pęcznienie w wysokiej temperaturze i tworzenie izolacyjnej warstwy ochronnej. Opisano najnowsze prace firm produkujących środki obniżające palność tworzyw oraz problemy związane z potencjalną toksycznością produktów spalania niektórych antypirenów.

Słowa kluczowe: środki uniepalniające, antypireny, dodatki do tworzyw sztucznych, palność

CHAPTER 14

TREE STRUCTURES AND ANALYSIS OF VARIANCE IN CLASSIFICATION OF PARAMETERS OF THE MACHINE SYSTEMS

Summary: The modified logical tree is a logical tree structure where logical values of variables are coded at the tree branches. Logical trees determine the importance rank of parameters of the given machine system: the variable in the root is the most important, the variable at the last level is the least important. The given logical function can be coded at many modified logical trees. We can distinguish the optimum logical trees – they have the minimum number of branches after all graphical simplifications. The tree classifier is a logical tree structure where logical values of variables are coded in the tree nodes. Many tree classifiers can be built for the given logical function. All of them have the same number of branches arranged at different numbers of levels. Analysis of variance can be applied as a statistic method of classification of machine system parameters. After its application, we define a factor (design /service parameter) strongly influencing the mean value of the criterion objective function. This paper contains comparison of the results of analyses obtained with the method of modified logical trees and tree classifiers, and the method of analysis of variance. A trial of cooperation of those methods was also presented. An impeller-propeller pump was used as an example.

Key words: discrete optimization, Boolean variable, criterion objective function

1. INTRODUCTION

The logical tree is a graphical presentation of a logical function written as a sum of products where each component (a path at the tree from the root to the final node) is realization of one solution of the design task, and each factor in the product (a branch at the tree) is the logical variable corresponding to the defined parameter of the designed object. The paper considers so-called modified logical trees. They differ from traditional trees because they can code different variables at one level. Each logical function can be coded at many modified logical trees. This group contains the optimum trees – they have the minimum number of branches after a graphical simplification. Classification of the machine system parameters at the optimum logical tree is defined by occurrence of variables at successive levels of the tree: the variable in the root is the most important, as the distance from the root increases, the importance rank of the root decreases.

The tree classifier is a logical tree structure where logical values of variables are coded in the tree nodes. Many logical variables can occur at the given level of the tree classifier. Many tree classifiers can be build for the given logical function. All of them have the same number of branches arranged at the different number of levels. Unlike the optimum logical trees, the classifier does not define the importance rank of the considered variables but it says which change of the numerical value of the machine system parameter influences behaviour of the machine system (a value of the criterion objective function).

The analysis of variance belongs to the basic devices of mathematical statistics. It can be successfully applied as the statistic method of classification of design / service parameters of the machine system. After application of the analysis of variance we can define a factor (a design/service parameter) strongly influencing the value of the criterion objective function. This paper presents comparison of the results of analyses obtained with the method of modified logical trees and tree classifiers, and the method of analysis of variance. Possibility of cooperation of these methods was also considered. The impeller-propeller pump was used as the example.

2. COMPARISON OF CLASSIFICTION METHODS

2.1. Description of the test propeller-type water turbine

Tree structures and analysis of variance are going to be applied for the impeller – propeller pump. The test turbine was designed and made with use of the design elements for the pump 25P21-2 produced in series by Warszawska Fabryka Pomp i Armatury WAFAPOMP S.A. The tests were performed for different angles of rotor blades φ [º], and at the test stand the following physical quantities were determined: n – rotational speed [1/min]; Q – capacity of water turbine [m^3/min]; N – power [kW]; H – difference of levels [m]; η - efficiency [%]. The obtained results of measurements (Table 1) were coded by logical variables in the following way: φ = 10 or 13 – logical variable takes the value **0**, φ = 17 or 21 – code **1**; n = 800 or 1000 – code **0**, n = 1200 or 1400 – code **1;** $Q \in (3;5,6]$ – code **0**, $Q \in (5,6;8]$ - code **1**; $H \in (4;7]$ –

code **0**, ***H*** ∈(7;10] – code **1**; ***N***∈(1;4] – code **0**, ***N***∈(4;7] – code **1.** The code notation of the Boolean presented in Table 2 was obtained.

Table 1

The results of measurements obtained for the test turbine

Lp	***φ***	***n***	***Q***	***H***	***N***	***η***
1	21	800	5,96	4,6	2,3	51,1
2	21	1000	7,77	8,5	5,4	50,0
3	21	1200	7,97	6,8	4,7	52,0
4	17	800	5,30	4,7	2,2	52,5
5	17	1000	6,40	5,7	3,2	53,3
6	17	1200	7,15	6,7	4,3	54,9
7	17	1400	7,93	7,9	5,4	52,9
8	13	800	4,63	4,3	1,9	58,8
9	13	1000	5,62	6,5	3,6	61,4
10	13	1200	6,30	7,7	4,9	63,6
11	13	1400	7,03	9,3	6,7	63,0
12	10	800	3,85	4,5	1,5	52,4
13	10	1000	5,09	7,3	3,5	57,0
14	10	1200	5,54	8,3	4,5	59,7
15	10	1400	5,79	8,2	4,7	63,1

Table 2

The code notation of logical function for measurements from Table 1

Lp	***φ***	***n***	***Q***	***H***	*N*	*η*
1	1	0	1	0	0	51,1
2	1	0	1	1	1	50,0
3	1	1	1	0	1	52,0
4	1	0	0	0	0	52,5
5	1	1	1	1	1	52,9
6	0	0	0	0	0	58,8
7	0	0	1	0	0	61,4
8	0	1	1	1	1	63,6
9	0	0	0	1	0	57,0
10	0	1	0	1	1	59,7

1.2. Application of one-factor analysis of variance

Each parameter from the group *φ, n*, *Q*, *H*, *N* was assumed as a factor influencing efficiency *η* (the criterion objective function). The Shapiro-Wilk test shows that at the significance level 0,05 suitable subgroups of data come from the normal population (Table 3). Homogeneity of the variance is also kept (Table 4). Thus, all the assumptions necessary for application of the analysis of variance are satisfied.

Table 3

Results of the Shapiro-Wilk test

	N	W	p
φ=0	5	0,947	0,715
φ=1	5	0,990	0,979
***n*=0**	6	0,925	0,539
***n*=1**	4	0,894	0,404
***Q*=0**	4	0,895	0,405
***Q*=1**	6	0,815	0,080
***H*=0**	5	0,840	0,165
***H*=1**	5	0,983	0,951
***N*=0**	5	0,944	0,696
***N*=1**	5	0,897	0,393

Table 4

Results of the Bartlett test

	Bartlett, chi-sq.	df	p
φ	1,938	1	0,164
n	0,121	1	0,728
Q	1,011	1	0,315
H	0,081	1	0,776
N	0,295	1	0,587

Table 5

Results of the one-factor analysis of variance

	F	df	p
φ	45,788	1	0,000
n	0,356	1	0,567
Q	0,324	1	0,585
H	0,217	1	0,654
N	0,026	1	0,875

The results obtained according to the one-factor analysis of variance in the package STATISTICA PL are presented in Table 5.

Results of the Fisher test are essential from the statistical point of view only when φ is the grouping variable. It means that at the significance level 0,05, the mean values of efficiency are different for different angles of blade settings (φ).

1.3. Application of tree structures

Structure of the optimum modified logical tree is based on the idea of the common path and possibility of forecast of so-called graphical simplifications on the tree. The applied logical function must be presented in both canonical alternate normal form (Table 2) and so-called reduced alternate normal form (Table 6).

Table 6

The reduced alternate normal form of the function from Table 2

Lp	*φ*	*n*	*Q*	*H*	*N*
1	-	0	-	0	0
2	0	0	0	-	0
3	1	1	1	-	1
4	0	1	-	1	1
5	1	-	1	1	1
6	-	1	1	1	1

After application of the algorithm for formulation of the optimum modified logical tree, the tree presented in Fig.1 was obtained.

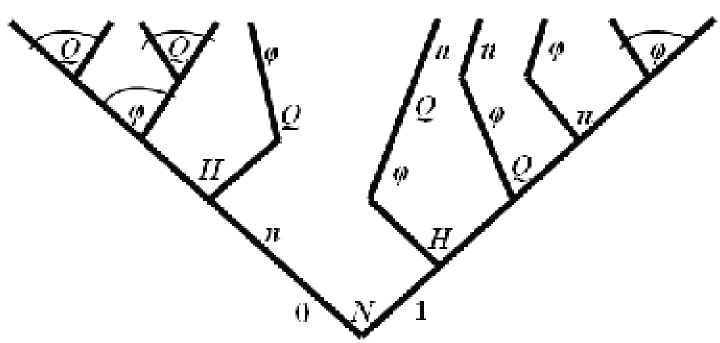

Fig. 1. The optimum modified logical tree for the function from Table 2

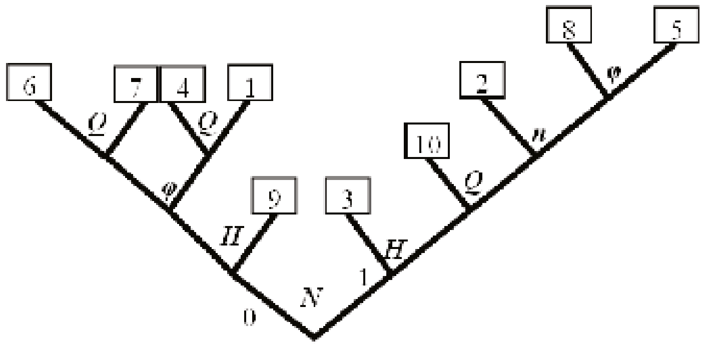

Fig. 2. The tree classifier corresponding to the tree from Fig. 1

The tree classifier shown in Fig.2 is a result of application of graphical operations of simplification and avoidance at the tree from Fig. 1.

1.4. Application of the results of analysis of variance for generation of the modified logical tree

While applying the one-factor analysis of variance, the angle of blade setting φ was assumed as the only parameter strongly distinguishing the average efficiency of the turbine. This statement can be applied as a guideline for formulation of the modified logical tree: let φ be in the root of the built tree. It appears that independently on the variables at the successive levels of that tree, the values of the tested parameters providing high efficiency of the turbine will be coded at the left side of the tree. At the right side of the tree, the values providing low efficiency will be coded (Fig. 3).

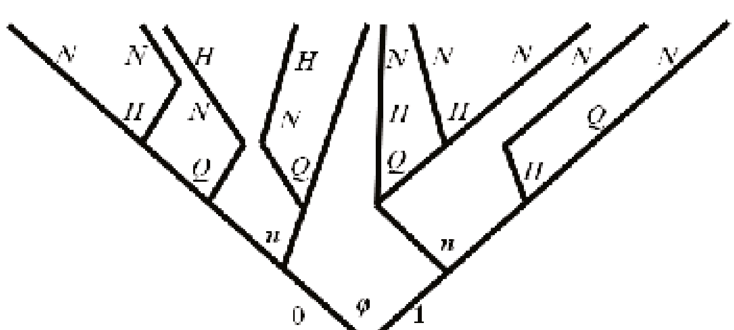

Fig. 3. The modified logical tree generated according to analysis of variance

1.5. Application of the results of analysis of variance for construction of the tree classifier

Results of the analysis of variance cause that the angle of blade setting φ has to be placed in the root of the constructed classifier. The data from Table 2 and operations of simplification and avoidance were applied for formulation of the analytic method of construction of the tree classifier for the considered case (1). The parameter subscript in equation (1) means its logical value.

$$\begin{gathered}\varphi_1(n_1(H_1Q_1N_1+H_0Q_1N_1)+n_0(Q_1(H_1N_1+H_0N_0)+Q_0H_0N_0))+\\+\varphi_0(n_1(Q_1H_1N_1+Q_0H_1N_1)+n_0(Q_1H_0N_0+Q_0(H_1N_0+H_0N_0)))=\\=\varphi_1(n_1(H_1+H_0)+n_0(Q_1(H_1+H_0)+Q_0))+\varphi_0(n_1(Q_1+Q_0)+n_0(Q_1+Q_0(H_1+H_0)))\equiv 1\end{gathered} \tag{1}$$

A graphical form of the classifier is presented in Fig. 4.

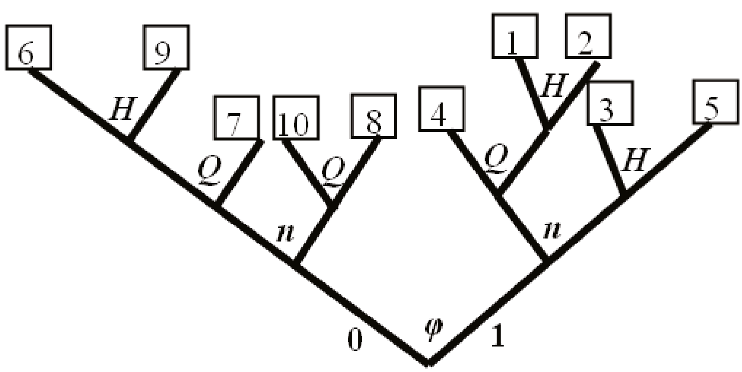

Fig. 4. The tree classifier corresponding to the tree from Fig. 3

3. CONCLUSIONS

The analysis of variance allows to determine the parameters strongly influencing the mean value of the criterion objective function (turbine efficiency). There are some design/service parameters of the impeller-propeller pump influencing its efficiency but only influence of the angle of blade setting φ is really important.

The optimum modified logical trees code changes of the machine system parameters providing the least possible number of changes on condition that

the system behaves in a particular way. The tree from Fig.1 codes values of the pump parameters with use of only 19 branches (after graphical simplifications) but with *N* in the root. It means that the minimum possible number of changes of numerical values of parameters of the tested machine system takes place when the changes start from the parameter *N*, and efficiency reaches values from the range [50,0%; 63,6%].
The classifier from Fig. 2 recognizes values of efficiency of the considered pump after checking numerical values of some (from 2 to 5) parameters. The final nodes – like on the tree from Fig. 1 – are not arranged from the point of view of the efficiency value.
Application of the results of analysis of variance for construction of the modified logical trees does not guarantee optimum trees having the minimum number of branches, but it provides classification of parameters of the designed machine system in respect of value of the criterion objective function. The result of one-factor analysis of variance shows that φ should be placed in the root of the modified logical tree. The tree from Fig.3 has 34 branches but it classifies the parameters very well, because the parameters providing good efficiency of the turbine (from 57,0% to 63,6%) are coded at the left side of the tree, and the right side contains the parameters giving efficiency from the lower ranges (from 50,0% to 52,9%). The classifier from Fig. 4 recognizes efficiencies of the impeller-propeller pump after checking 3 or 4 numerical values of the tested parameters. Moreover, the final nodes are arranged: at the left side the classifier recognizes high efficiencies, and at the right side – low efficiencies.

The presented different methods can cooperate. Cooperation of analysis of variance and tree structure methods can be extended to the multiple-valued case.

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STRUKTURY DRZEWIASTE I ANALIZA WARIANCJI W KLASYFIKACJI PARAMETRÓW UKŁADU MASZYNOWEGO

Drzewa logiczne ustalają rangę ważności parametrów danego układu maszynowego: zmienna w korzeniu jest najważniejsza, zmienna na ostatnim piętrze jest najmniej ważna. Klasyfikator nie ustala rangi ważności analizowanych zmiennych, ale wskazuje jaka zmiana wartości liczbowej parametru układu maszynowego wpłynie na zachowanie się tego układu (wartość kryterialnej funkcji celu). Analiza wariancji wskazuje czynnik (parametr konstrukcyjny/eksploatacyjny), który istotnie wpływa na wartość średnią kryterialnej funkcji celu.

Na przykładzie pompy wirowej śmigłowej, przedstawiono porównanie wyników analiz uzyskanych metodą zmodyfikowanych drzew logicznych, klasyfikatorów drzewiastych oraz metodą analizy wariancji, a także pokazano próbę współdziałania tych metod.

Słowa kluczowe: optymalizacja dyskretna, zmienna boolowska, kryterialna funkcja celu.

CHAPTER 15

INFLUENCE OF VALENCE OF THE PARAMETERS OF THE CRITERIAL OBJECTIVE FUNCTION IN THE MULTIPLE-VALUED NETWORK-TREE METHOD

Minimization of the Boolean function is used for search the simplest function equivalent to the given initial function, i.e. the function that can be written with the minimum number of logical symbols (negation, conjunction, alternative).
Minimization of two-valued logical functions is performed with different methods, such as the network-tree method, a result of which is obtained by transformation of suitable figures. In this method, all the graphical transformations are equivalents in the sense of two-valued transformations of the given logical function.
The multiple-valued network-tree method of minimization of functions can be used for variable values in the criterial objective functions described in a multiple-valued way with suitable coding. The selected variables are coded as multiple-valued variables, and not as complex systems of two-valued variables.

Key words: minimization of logical functions, network-tree method

1. INTRODUCTION

Optimisation performed according to the network method requires elaboration of a set of elements which have to be subjected to minimization. If we have a given space of possible acceptable solutions satisfying conditions of the design task, we can use optimisation of these solutions. In the case when the given design system is described by the function of design variables and the value of each variable cannot be changed in a continuous way, then the area of acceptable solutions has a discrete structure. Minimization of two- and multiple-valued logical functions can be performed in an analytical, numerical or graphical way. The network-tree method based on the graphical analysis of a Boolean or multiple-valued function is a method allowing for a fast optimisation. In the design process, logical decision trees with no isolated branches play an important role, because design guidelines must be always described with all the decision structure, such as the canonical and minimum alternate normal form of the logical function. If the traditional logical tree is drawn on the rectangular network, geometrical properties can be additionally used in the minimization process.

2. THE TRADITIONAL NETWORK-TREE METHOD

Presentation of the multiple-valued logical function written in both canonical alternate normal form and in a graphical way (as a logical tree) can be traditional, where a notation of the function is used with a suitable value number for the given variable, or modified as a system composed of two-valued logical functions. Such presentation allows to consider the multiple-valued logical function in the approach of the methods applied for two-valued logical functions, however such a presentation must be understood as a suitable system of two-valued functions.

2.1. Position of the logical tree on the network

The logical tree is a graphical image of a selected Boolean function. Analysis of the logical tree according to the network-tree method proceeds on a plane network where the tree is suitably coded.

EXAMPLE

The following logical tree corresponds to the Boolean function $f(x_1, x_2, x_3)$ written in the canonical alternate normal form $f(x_1, x_2, x_3) = 000, 100, 010, 110, 111$ drawn on the network

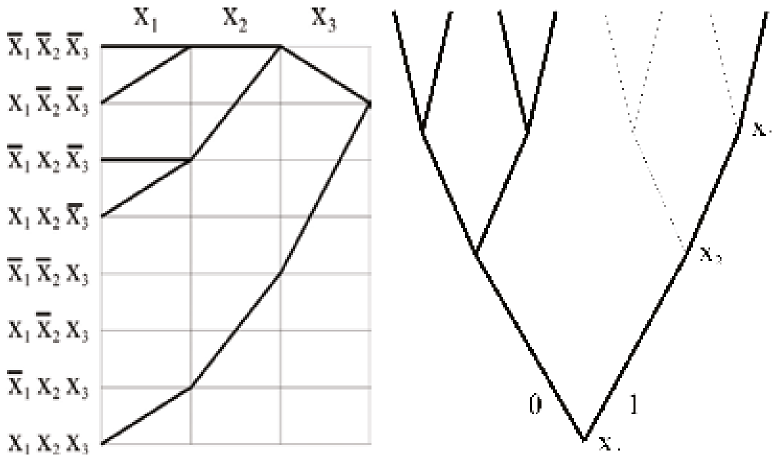

Fig.1. Images of the function *f(x₁, x₂, x₃)* on the network and the logical tree

In the case of multiple-valued logical functions, it is not possible to present the tree according to the network-tree method because of the two-dimensional character of the network where the tree is located. However, it is possible to present each variable of the multiple-valued logical function as a system of two-valued logical functions composed of zero or one. Thus, for the multiple-valued function, each variable must be re-coded for the system (sequence) compound of only zero or one.

In the case of the function where the variables are three-valued, the values 0, 1, 2 can be re-coded for 00, 01, 10 respectively, according to the method of a change of a number for the binary system, because the state 11 is empty.

EXAMPLE

For the function $f(x_1, x_2, x_3)$, written in the canonical alternate normal form: $f(x_1, x_2, x_3) = 010, 102, 101, 220, 201$, each variable should be re-coded according to the above notation.

Table 1.

Values of the function $\boldsymbol{f(x_1, x_2, x_3)}$ before and after coding

x_1	x_2	x_3	x_1	x_2	x_3
0	1	0	00	01	00
1	0	2	01	00	10
1	0	1	01	00	01
2	2	0	10	10	00
2	0	1	10	00	01

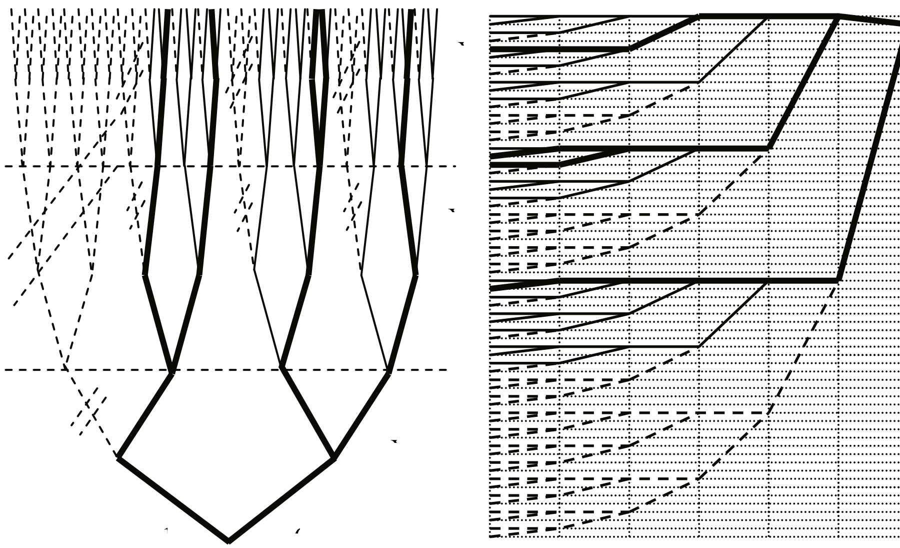

Fig. 2. Graphical image of the system of two-valued logical functions

2.2. Minimization of the function with the network-tree method

Minimization of the Boolean function is used for search the simplest function equivalent to the given initial function, i.e. the function that can be written with the minimum number of letters and logical symbols (negation, conjunction and alternative).

Minimization of two-valued logical functions can be performed according to a special algorithm, i.e. the network-tree method. Its result is obtained by transformation of suitable figures. All the graphical transformations in the network-tree method are equivalents in the sense of logical transformations of the given logical function. .

Minimization of the logical function from the graphical point of view allows to search the tree with the minimum number of branches as well as to search the system of variables and determine the priority of their changes. Having the determined importance rank of logical variables, we can define which variable is the least important, intermediately important or the most important. Depending on the set of data and possibilities of suitable re-coding that set for the two- or multiple-valued system, while determining the importance rank of variables we can give the method of selection of sequence of suitable variable changes.

3. THE NETWORK-TREE METHOD FOR THE MULTIPLE-VALUED LOGICAL FUNCTION

3.1. Analysis of values of the parameters

The multiple-valued network-tree method of function minimization can be applied for variable values in the criterial objective functions described in the multiple-valued way with suitable coding. The chosen variables are coded as multiple-valued variables, and not complex systems of two-valued variables.

For an exemplary three-valued variable x_1 taking the values 0, 1, 2, coding of the function can be drawn on the rectangular network in the following way:

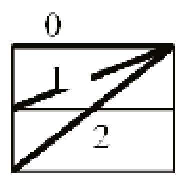

Fig. 3. Coding for an exemplary variable

Coding performs on two squares, at each vertex respectively. In the case of multiple-valued function, coding on the network requires as much place as valence of that function. .

3.2. Tree position on the network

Using such coding, it is possible to draw a logical tree on the plane network, keeping the same number of levels at the logical tree for the determined number of variables.

Table 2 presents an universal method of drawing the multiple-valued function $f(x_1, x_2, ..., x_n)$ on the plane network.

Table 2

Algorithm of drawing the multiple-valued function on the plane network

Valence of function	**Number of squares for one variable**	**Position of the next variable**		**Network size**
1	0	0	0	0
2	1	2; 4; 6; 8;...	2*lz	2^{lz}-1
3	2	3; 6; 9; 12; 15; 18;21;...	3*lz	3^{lz}-1
4	3	4; 8; 12; 16; 20; 24; 28;...	4*lz	4^{lz}-1
5	4	5; 10; 15; 20; 25; ...	5*lz	5^{lz}-1
...	...	...	...	...
w	**w-1**	**1*w; 2*w; 3*w; ...**	**w*lz**	**w^{lz}-1**

where :

- **w** –valence of the function $\boldsymbol{f(x_1, x_2, ..., x_n)}$;

- **lz** – number of variables of the given function $\boldsymbol{f(x_1, x_2, ..., x_n)}$;

Each line of the network corresponds to a number from **0** to **w^{lz}-1.** This number can be expressed by valence of variables.

Each number is changed into a sequence of digits corresponding to the change into a suitable valence of the given variables.

EXAMPLE:

For the function $f(x_1, x_2, x_3)$ where each variable takes values from the range 0, 1, 2, the notation is as follows:

Tabela 3.

The chosen values for the function $f(x_1, x_2, x_3)$

Lp.	X_1	X_2	X_3
1	0	1	1
2	2	0	1
3	1	0	1
4	2	2	0
5	0	0	1
6	1	2	0
7	0	2	0
8	2	1	0
9	1	1	0
10	0	1	0

The logical tree situated on the network is shown in Fig.4.

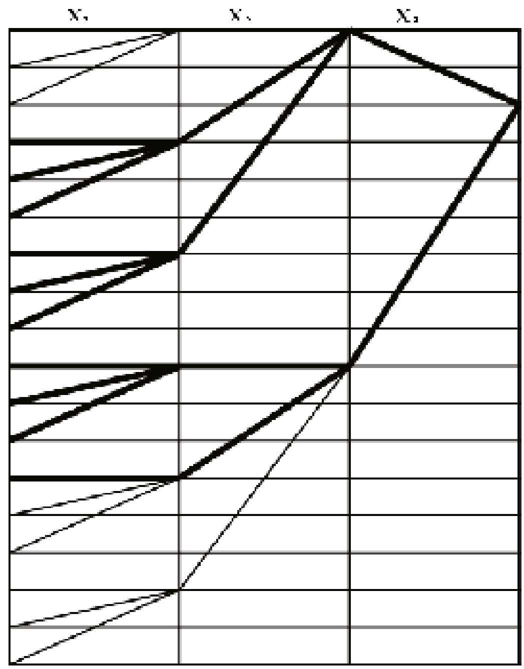

Fig. 4. The logical tree on the network for the system of variables x_1, x_2, x_3.

In the network-tree method for the two-valued logical function, it is possible to code nodes of the tree situated on the network. This property is also valid for the multiple-valued logical function. Each node for the system of logical trees corresponds to one decimal number coming from the read-out of each extended product of the canonical alternate normal form from the left to the right and the change into the decimal system.

4. CONCLUSIONS

Minimization of the Boolean function is used for search of the simplest function equivalent to the given initial function, i.e. the function that can be written with the least number of letters and logical symbols (negation, conjunction, alternative). A number of iterations and – in a consequence – a rate of the applied algorithm are of a great importance in the chosen method.

Minimization of the two-valued logical functions can be performed with different methods. The modified Quine-McCluskey algorithm is one of such methods. There are also graphical methods, such as the network-tree method, where the results are obtained after transformation of suitable figures. All the graphical

transformations in the network-tree method are equivalents in the sense of two-valued transformations of the given logical function. Thus, valence of variables is very important. In the multiple-valued network-tree method, it is not necessary to code the multiple-valued variables into two-valued variables, and in a consequence the algorithm based on this method includes a less number of iteration.
The network-tree method in traditional and modified forms can be applied in methodology of machine system designing.

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WPŁYW WARTOŚCIOWOŚCI PARAMETRÓW KRYTERIALNEJ FUNKCJI CELU W WIELOWARTOŚCIOWEJ METODZIE SIATKOWO-DRZEWIASTEJ

Proces minimalizacji funkcji boolowskiej służy do znalezienia najprostszej funkcji równoważnej danej funkcji początkowej, to znaczy takiej, która może być zapisana za pomocą najmniejszej liczby liter i symboli logicznych (negacja, koniunkcja, alternatywa).

Minimalizację dwuwartościowych funkcji logicznych prowadzi się różnymi metodami jak np. metoda siatkowo-drzewiasta, której wynik otrzymuje się przekształcając odpowiednie rysunki. Wszystkie przekształcenia graficzne w metodzie siatkowo-drzewiastej są odpowiednikami równoważnościowymi w sensie przekształceń dwuwartościowych danej funkcji logicznej.

Dla wartości zmiennych w kryterialnej funkcji celu opisanych wielowartościowo z odpowiednim zakodowaniem można zastosować wielowartościową metodę siatkowo – drzewiastą minimalizacji funkcji. Wybrane zmienne kodowane są jako zmienne wielowartościowe, a nie układy piętrowe zmiennych dwuwartościowych..

Słowa kluczowe: minimalizacja funkcji logicznych, metoda siatkowo-drzewiasta

CHAPTER 16

THE UNIFORM METHOD OF OBTAINING INTEGRAL INEQUALITIES

The uniform method of obtaining various types of integral inequalities for the first time was given by Florkiewicz and Rybarski in [6] to examine the Sturm-Liouville type inequalities.Then it was extended to different types of integral inequalities of the first and second order. Also Hardy type inequality of the third order was derived by it.

Our aim is to describe the method and the inequalities it allows to obtain.

Next we compare the method with the other ones and show the difference in results.

Keywords: integral inequality, absolutely continuous function

1. THE UNIFORM METHOD OF OBTAINING INTEGRAL INEQUALITIES

1.1. Introduction

Integral inequalities is a branch of mathematics developing rapidly during last years. It has applications both to other branches of mathematics and to other areas. Many authors study different types of integral inequalities and use different methods to obtain them. We refer the reader to the monographs by Kufner, Person [12] and Mitrinović, Pečarić, Fink [15]. One of the methods is a uniform method of obtaining different types of integral inequalities introduced by Florkiewicz and Rybarski in [6]. The authors obtained the Sturm-Liouville type inequalities by this method. For a positive weight function r and an auxiliary function φ they defined the weight function s such as a suitable differential identity was satisfied. Next this identity was used to to construct the class$H$ of functions for which the inequality

$$\limsup_{t\to\beta} vh^2 - \liminf_{t\to\alpha} vh^2 \le \int_I (rh'^2 - sh^2)dt$$

holds.

It appeared that the method can be modified to obtain different types inequalities of the first and second order. Also Hardy type inequality of the third order was derived by it.

In this monograph we describe the method and give the inequalities it allowed obtain.

We will also compare the method with the other ones and show the differences in results.

1.2. Notations

Let $I = (\alpha, \beta), -\infty \leq \alpha < \beta \leq \infty,$ be an arbitrary open interval.

Let $AC(I)$ denote the class of real functions f defined and absolutely continuous on the interval I, and $AC^1(I)$ the class of functions $f \in AC(I)$ such that $f' \in AC(I)$.

Let $\mathrm{A} = \mathrm{A}(r, \varphi)$ be a class of functions such that r is a weight function positive and absolutely continuous on I and φ is an arbitrary function positive and absolutely continuous on I, e.g. $r > 0, r \in AC(I),$ and $\varphi > 0, \varphi \in AC(I).$

Let $\widetilde{\mathrm{A}} = \widetilde{\mathrm{A}}(r, \varphi)$ be a class of functions such that r is a weight function positive and absolutely continuous on I and φ is an arbitrary function positive and absolutely continuous on I, e.g. $r > 0, r \in AC(I),$ and $\varphi > 0, \varphi \in AC^1(I).$

1.3. The method

In [6] the uniform method of obtaining integral inequalities was introduced for the first time to obtain the Sturm-Liouville integral inequalities. Namely for $(r, \varphi) \in \mathrm{A}$ the functions $s = -(r\varphi')'\varphi^{-1}$ and $v = r\varphi'\varphi^{-1}$ were defined. For such functions *s* and *v* the identity

$$rh'^2 = sh^2 + (vh^2)' + r\varphi^2(h\varphi^{-1})'^2 \tag{1}$$

is valid.

Let H be the class of functions h absolutely continuous on the interval I satisfying following integral and limit conditions

$$\int_I rh'^2 dt < \infty, \quad \int_I sh^2 dt > -\infty, \tag{2}$$

$$\liminf_{t\to\alpha} vh^2 < \infty, \quad \limsup_{t\to\beta} vh^2 > -\infty. \tag{3}$$

For such functions h the identity was proved:

$$\int_I rh'^2 dt = \int_I sh^2 dt + \int_I r\varphi^2 (h\varphi^{-1})'^2 dt + \lim_{t\to\beta} vh^2 - \lim_{t\to\alpha} vh^2 \tag{4}$$

Using this identity the following theorem holds true:

Theorem 1. *Let r and* φ *be in* A. *If* $h \in H$, *then*

$$\limsup_{t\to\beta} vh^2 - \liminf_{t\to\alpha} vh^2 \le \int_I (rh'^2 - sh^2)dt. \tag{5}$$

If additionally $\liminf_{t\to\alpha} vh^2 \le 0$ and $\limsup_{t\to\beta} vh^2 \ge 0$ the inequality:

$$\int_I sh^2 dt \le \int_I rh'^2 dt, \tag{6}$$

is valid.

1.4. First order integral inequalities

Further research allowed to obtain, using the uniform method of obtaining the inequalities, the Hardy type, the Weyl type, the Opial type and some quadratic integral inequalities of the first order .

In [2] Florkiewicz, using the uniform method, generalized the inequalities (4) and (5) for the case of $p > 1$. For given $(r,\varphi) \in \mathrm{A}$, $r\,|\,\varphi'\,|^{p-1}\,\mathrm{sgn}\,\varphi' \in AC(I)$ the functions *s* and *v* were modified so that $s = -(r\,|\,\varphi'\,|^{p-1}\,\mathrm{sgn}\,\varphi')'\varphi^{1-p}$, $v = r\,|\,\varphi'\,|^{p-1}\,\mathrm{sgn}\,\varphi'\varphi^{1-p}$ and inequality

$$\int_I s\,|\,h\,|^p\,dt + \lim_{t\to\beta} v\,|\,h\,|^p - \lim_{t\to\alpha} v\,|\,h\,|^p \le \int_I r\,|\,h'\,|^p\,dt \tag{7}$$

was proved for $h \in AC(I)$ such that $\int_I r\,|\,h'\,|^p\,dt < \infty$, $\int_I s\,|\,h\,|^p\,dt > -\infty$ and $\liminf_{t\to\alpha} v\,|\,h\,|^p < \infty$, $\limsup_{t\to\beta} v\,|\,h\,|^p > -\infty$. If $\liminf_{t\to\alpha} v\,|\,h\,|^p \le \limsup_{t\to\beta} v\,|\,h\,|^p$, then the Hardy type inequality

$$\int_I s\,|\,h\,|^p\,dt \le \int_I r\,|\,h'\,|^p\,dt \tag{8}$$

holds.

Further modifications of the method allowed to obtain in [3] the Weyl type. For appropriate functions $(r,\varphi) \in \mathrm{A}$, two weight functions $s = r\,|\,\varphi\,|^p$, $u = (r\,|\,\varphi\,|^{p-1}\,\mathrm{sgn}\,\varphi)'$, $p > 1$ were defined and the wide class of functions *h* for which inequality holds was described.

$$\int_I u\,|\,h\,|^p\,dt \le p\,\left(\int_I r\,|\,h'\,|^p\,dt\right)^{\frac{1}{p}}\left(\int_I s\,|\,h\,|^p\,dt\right)^{\frac{1}{q}}, \quad q = \frac{p}{p-1}, \tag{9}$$

In [11] the Opial type inequalities were obtained. In earlier papers to get the inequality the weight functions were calculated explicitly. In this case $s \in AC(I)$ is given as a solution of differential inequality $\frac{1}{2}s' - (r\varphi')'\varphi^{-1} \geq 0$. Taking $v = -\frac{1}{2}s + r\varphi'\varphi^{-1}$, the class of function $h \in AC(I)$ satisfying the inequality

$$\lim_{t\to\beta} vh^2 - \lim_{t\to\alpha} vh^2 \leq \int_I (rh'^2 - shh')dt \tag{10}$$

was constructed ($\int_I rh'^2 dt < \infty$, $\int_I shh' dt > -\infty$, $\liminf_{t\to\alpha} vh^2 < \infty$, and $\limsup_{t\to\beta} vh^2 > -\infty$).

Next, depending on r, φ, the Opial type integral inequalities:

$$\int_I s\,|hh'|\,dt \leq \int_I rh'^2\,dt \tag{11}$$

were given.

This result was extended in [4] to inequalities of the form

$$\int_I s\,|h|^p|h'|\,dt \leq \int_I r\,|h'|^{p+1}\,dt, \quad p > 0. \tag{12}$$

In [5] to get quadratic integral inequalities

$$\int_I (rh'^2 + 2shh' + uh^2)dt \geq 0 \tag{13}$$

two weight functions were defined. Similiary to Opial type, s and u satisfy the differential inequality $s' - u + (r\varphi')'\varphi^{-1} \leq 0$ for given r, φ. For such weights the class H of functions $h \in AC^1(I)$ satisfying the inequality (13) is constructed.

In all this papers the classes of functions satisfying some integral and limit conditions, for which different types of integral inequalities holds were given. They depended on positive and absolutely continuous functions r and φ. Then functions for which the inequalities hold were characterized by boundary conditions in order to simplify limit conditions.

1.4. Second order integral inequalities of Hardy type

Further studies allowed to extend, in [7], the method to the second order integral inequalities of Hardy type. For given $(r, \varphi) \in \widetilde{A}$, such that $r\varphi'' \in AC^1(I)$ and $\varphi'' \leq 0$ a.e. on I. the weight function $s = (r\varphi'')''\varphi^{-1}$ was defined.

Let H denote the class of functions $h \in AC^1(I)$ satisfying the conditions:

$$\int_I rh''^2 dt < \infty \quad , \quad \int_I sh^2 dt > -\infty \qquad (14)$$

$$\liminf_{t\to\alpha} S(t,h,h') < \infty \quad , \quad \limsup_{t\to\beta} S(t,h,h') > -\infty \qquad (15)$$

and

$$\liminf_{t\to\alpha} S(t,h,h') \le \limsup_{t\to\beta} S(t,h,h'), \qquad (16)$$

where $S(t,h,h') = v_0 h^2 + 2v_1 hh'$, with $v_0 = r\varphi''(\varphi^{-1})' - (r\varphi'')'\varphi^{-1}$, $v_1 = r\varphi''\varphi^{-1}$.

Then the following theorem holds true

Theorem 2. *For every function $h \in H$ the inequality*

$$\int_I sh^2 dt \le \int_I rh''^2 dt \qquad (17)$$

holds.

If $\varphi'' \not\equiv 0$ and $h \not\equiv 0$ then the inequality (17) becomes an equality if and only if $h = c\varphi$ with $c =$ const $\neq 0$ and $\varphi \in H$, $\lim_{t\to\alpha}(v_0\varphi^2 + 2v_1\varphi\varphi') = \lim_{t\to\beta}(v_0\varphi^2 + 2v_1\varphi\varphi')$.

To prove Theorem 2 the Picone'a identity

$$rh''^2 = sh^2 + (v_0 h^2 + v_1 hh')' + g \qquad (18)$$

was shown with $g = r[\varphi(h\varphi^{-1})'' + 2\varphi'(h\varphi^{-1})']^2 - 2r\varphi\varphi''[(h\varphi^{-1})']^2$.

Next different types of the boundary points α and β were studied depending on v_0 and v_1, what allowed to simplify the limit conditions (15), (16) for the class *H*.

Then some examples were considered. It was shown that some 'standard' Hardy integral inequality given in [12] can be also obtained with $m = 2$ and $p = q = 2$. For $r = t^a$ and $\varphi = t^{\frac{3-a}{2}}$ on $I = (0,\infty)$, where $1 < a < 3$ is an arbitrary constant, the inequality

$$\left[\frac{(a-1)(a-3)}{4}\right]^2 \int_0^\infty t^{a-4} h^2 dt < \int_0^\infty t^a h''^2 dt \qquad (19)$$

holds for every function $h \in AC^1((0,\infty))$ satisfying the conditions: $\int_0^\infty t^a h''^2 dt < \infty$,

$$-\infty < \lim_{t\to 0+} S(t,h,h') \le \lim_{t\to\infty} S(t,h,h') < \infty, \text{ where}$$

$$S(t,h,h') = t^{a-3}h^2 - 2t^{a-2}hh'.$$

Another important example is the case with Chebyshev weight functions. It was shown for $r=(1-t^2)^a$, $\varphi=(1-t^2)^{2-a}$ on I, where $1\le a<\frac{3}{2}$ is an arbitrary constant, that

$$4(a-2)(2a-3)\int_{-1}^{1}(1-t^2)^{a-2}h^2dt \le \int_{-1}^{1}(1-t^2)^a h''^2 dt \quad (20)$$

holds for $h\in AC^1((-1,1))$ satisfying the integral condition

$$\int_{-1}^{1}(1-t^2)^a h''^2 dt < \infty \quad (21)$$

and the limit conditions $h(-1)=(hh')(-1)=h(1)=(hh')(1)=0$ in case of $a=1$ or $h(-1)=h'(-1)=h(1)=h'(1)=0$ if $1<a<\frac{3}{2}$.

Modifications of the identity (18) allowed to obtain, in [8], the inequality (17) in essentially different class $\widetilde{H}$.

For $(r,\varphi)\in\widetilde{\mathrm{A}}$, such that $r\varphi''\in AC^1(I)$ the condition $\varphi''\le 0$ was exchanged with $w=(r\varphi')'\varphi+2r\varphi\varphi''-2r\varphi'^2\le 0$. The class $\widetilde{H}$ of functions $h\in AC^1(I)$ satisfy the conditions (14)-(16) but $S(t,h,h') = w_0h^2+2w_1hh'+w_2h'^2$,

$w_0 = r(\varphi^{-1}\varphi')^3 + r\varphi''(\varphi^{-1})' - (r\varphi'')'\varphi^{-1}$, $w_1 = r(\varphi^{-1}\varphi')'$, $w_2 = r\varphi^{-1}\varphi'$.

The difference between H and $\widetilde{H}$ was shown on examples. The 'standard' Hardy inequality (19) couldnot be obtained in [7] for $a<1$ and $a>3$ as the condition $\varphi''<0$ was not satisfied. In [8] the condition $\varphi''<0$ was replaced by $w\le 0$, and therefore (see [18]) for $a<1$ and $a>3$, the inequality (19) holds for all functions h such that $\int_0^\infty t^a h''^2 dt<\infty$ and $-\infty < \lim_{t\to 0+} P(t,h,h') \le \lim_{t\to\infty} P(t,h,h') < \infty$,

where $P(t,h,h') = \frac{3-a}{2}\left[t^{a-3}h^2 - 2t^{a-2}hh' + t^{a-1}h'^2\right]$

Similarly for Chebyshev weights the inequality (20) was obtained in [7] for $1 \le a < \frac{3}{2}$. In [8] the assumption $w \le 0$ on $(-1,1)$ instead of $\varphi'' \le 0$ on $(-1,1)$ allowed to it for $a < 1$ and functions h satisfying integral condition (21) and the limit conditions $h(-1) = h'(-1) = h(1) = h'(1) = 0$. Inequalities with Chebyshev weights for $a < 1$ are important because of applications (for example in probability).

Also the inequalities of the third order were obtained by this method. In [16] Muminov for given positive functions $r \in AC^1(I)$ and $\varphi \in AC^2(I)$ satisfying some additional assumptions put the weight function $s = (r\varphi''')'''\varphi^{-1}$ and has shown the following inequality

$$\int_I sh^2 dt \le \int_I rh'''^2 dt$$

for the class of functions $h \in AC^1(I)$ satisfying some integral and limit conditions.

Nowadays many authors consider the inequality of the form (17) e.g. Nasyrova and Stepanov in [17] . Further references can be found in [12] and [7].

Also Hardy inequalities of k -th order

$$\left(\int_\alpha^\beta s\,|\,h\,|^p\;dt\right)^{1/p} \le C\left(\int_\alpha^\beta r\left|h^{(m)}\right|^q\right)^{1/q} dt,$$

are considered, where m is given constant $m \ge 2$, $1 < p \le q < \infty$, r and s are nonnegative weight functions, $C = const$, (a,b) is an interval finished or not.

Hardy inequality was extended in different directions. Not only to higher order integral inequality, but also to the form

$$\left(\int_a^b \big(F(x)\big)^q u(x)dx\right)^{\frac{1}{q}} \le C\left(\int_a^b (f(x))^p v(x)dx\right)^{\frac{1}{p}} \qquad (24)$$

for $-\infty \le a < b \le \infty$, u, v - weight functions, $1 \le p \le \infty$, $0 < q \le \infty$ and $F(x) = \int_a^x f(t)dt$.

If $(Hf)(x) = \int_a^x f(t)dt$ denotes the Hardy operator, then the above inequality can be rewritten as

$$\| Hf \|_{q,u} \le C \| f \|_{p,v} \,.$$

Different authors considered weights u and v for which Hardy operator $(Hf)(x)=\int_a^x f(t)dt$ is bounded from L^p to L^q. Also the N-dimensional Hardy operator was considered (see [8]) and Hardy operator between normed spaces (see e.g. [10]).

However the method used by others is different and, in consequence, the result is different to the one obtained by the uniform method.

1.4. Quadratic second order integral inequalities

In [9], some quadratic integral inequalities of the second order were introduced.

Let r and u be weight functions positive and absolutely continuous on I and φ be an arbitrary function positive and absolutely continuous on I, (e.g. $(r,\varphi)\in\widetilde{\mathrm{A}},\ u>0,\ u\in AC(I)$). Put the weight function $s=-[(r\varphi'')''+(u\varphi')']\varphi^{-1}$. For such defined weihgts the class H of functions satisfying the inequality was constructed in the similar way as in case of Hardy type inequalities.

Then the following inequality holds true:

$$\int_I uh'^2\,dt\le\int_I(sh^2+rh''^2)dt. \tag{22}$$

Also for this inequalities some examples with Chebyshev weight functions were described. It was shown that for arbitrary constants $1\le a<\dfrac{3}{2}$ and $2(3-2a)<A\le(2-a)(1+2a)$, the inequality

$$A\int_{-1}^{1}(1-t^2)^{a-1}h'^2\,dt<(2-a)(A+4a-6)\int_{-1}^{1}(1-t^2)^{a-2}h^2dt+\int_{-1}^{1}(1-t^2)^{a}h''^2dt \tag{23}$$

holds for $h\in AC^1((-1,1))$ satisfying the conditions:

$$\int_{-1}^{1}(1-t^2)^{a}h''^2dt<\infty,\ \int_{-1}^{1}(1-t^2)^{a-2}h^2dt<\infty,$$
$$h(-1)=h'(-1)=h(1)=h'(1)=0 \tag{24}$$

Also in this case modifications of the method gave, in [20], the inequality (22) in essentially different class of functions. The difference between them was shown on the example with Chebyshev weights. The inequality (23) was studied in [20] with $a<1$ for functions h satisfying (24).

Unfortunately, the classical Hardy Littlewood, Polya's inequality (HLP) could not be obtained in this way.

In [19] the method was extended and the quadratic inequality (22) was obtained in more general class of functions $\widehat{H}$.

Let $\hat{A}$ denote the class of triples (r,φ_1,φ_2) of given functions $r>0,\ r\in AC(I),\ \ \varphi_1,\varphi_2\in AC^2(I)$. We require $\varphi_1\varphi_2'-\varphi_1'\varphi_2>0$. For given $(r,\varphi_1,\varphi_2)\in\hat{A}$ put the weight functions $u=-[(r\sigma')'-2r\tau]\sigma^{-1}$, $s=[(r\tau')'\sigma^{-1}]'+r\tau^2\sigma^{-2}$, with $\sigma=\varphi_1\varphi_2'-\varphi_1'\varphi_2$, $\tau=\varphi_1'\varphi_2''-\varphi_1''\varphi_2'$. For such defined weights and φ_1,φ_2 in the similar way as before the class $\widehat{H}$ of functions satisfying the inequality (22) was described by certain limit and integral conditions. Also in this case the limit conditions for the class $\widehat{H}$ were simplified (Theorem 10 in [19]).

It appeared that this modifications allowed to obtain the classical Hardy Littlewood, Polya's inequality (HLP) as a special example. Namely for $I=(0,\infty)$ and $0<a<b$ it was shown that the inequality

$$\int_0^\infty 2(b^2-a^2)h'^2dt\le\int_0^\infty((a^2+b^2)h^2+h''^2)dt \tag{25}$$

is valid for h satisfying integral

conditions $\int_0^\infty h''^2dt<\infty,\ \int_0^\infty (a^2+b^2)h^2dt<\infty$ with no limit conditions for

$0<a<b\le a\sqrt{3}$ and the limit condition $hh'(0)\le 0\ \ b>a\sqrt{3}.$

This result shows that the inequality (25) considered in[13] with classical HLP inequality for can be obtained as a special case from quadratic inequality of the second order (22) obtained by unform method with more general weights.

Also in this case some Chebyshev type inequalities were considered for $1\le a<\frac{5}{3}$

$$2(5-3a)\int_{-1}^1(1-t^2)^{a-1}h'^2\,dt<4(a-2)^2\int_{-1}^1(1-t^2)^{a-2}h^2dt+\int_{-1}^1(1-t^2)^a h''^2dt \tag{17}$$

holds for $h\in AC^1((-1,1))$ satisfying the conditions:

$$\int_{-1}^1(1-t^2)^a h''^2dt<\infty,\ \int_{-1}^1(1-t^2)^{a-2}h^2dt<\infty,$$
$$h(-1)=h'(-1)=h(1)=h'(1)=0.$$

The inequalities of the form (22) have been considered by others (see e.g. [14]) but different approach was used. Further detailed studies can be found in the monographs [13], [15].

1.5. A difference between other methods and the uniform of obtaining integral inequalities

As it was mentioned in 1.3 and 1.4 these types of inequalities has been also studied by others, but the methods that were used were different.

There are two main differences. Firstly in the uniform method to derive inequalities the identity is considered. Therefore equality conditions are obtained in natural way. It has a great importance in applications, as that way allows to find the best constant for inequality.Secondly because of the way of obtaining the inequality some assumptions on weights sufficient in other methods can be omitted. For example for Hardy type inequalities of higher order derived in different ways authors consider functions h vanishing together with some of their derivatives respectively up to the order $m-1$ either on one or on the both endpoints of (a,b). For such functions conditions on the weight functions r and s such that the inequality is fulfilled are found. In the uniform method given positive functions r and auxiliary φ the weight function s is determined directly and class of functions satisfying some integral and limit conditions for which the Hardy inequality (17) holds is built. The results obtained by others and by the uniform method do not cover. The method allows to omit some convergence assumptions on weight functions r and s, which are put by other authors, e.g. for an inequality with Chebyshev weights for $a<1$ in the uniform method functions r and s do not need to be integrable on $(-1,1)$. This is important, for example, in applications of integral inequalities to probability.

The importance of the method has been noticed as the method was described in a monograph [15].

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JEDNOLITA METODA OTRZYMYWANIA NIERÓWNOŚCI CAŁKOWYCH

W 1976 roku B. Florkiewicz i A. Rybarski w pracy: "Some integral inequalities of Sturm-Liouville type", *Colloq. Math.* **36** (1976), 127-141 opracowali jednolia metode otrzymywania nierówności całkowych. Metoda ta pozwoliła na otrzymywanie różnych typów nierówności całkowych pierwszego i drugiego rzędu na prostej rzeczywistej w różnych klasach funkcji.

W dniu dzisiejszym badane są nierówności wyższych rzędów, rzędu ułamkowego, tzw „fractional order" , a także nierówności w przestrzeniach R^n, jednak metody ich otrzymywania, a także wyniki są istotnie różne od otrzymanych przy pomocy jednolitej metody. Autorzy posługują się bowiem metodami, które ograniczają otrzymanie równości, co jest naturalnym efektem przy wykorzystaniu metody ich otrzymywania poprzez odpowiednie tożsamości wprowadzonej przez Florkiewicza i Rybarskiego. Metoda pozwala też ominąć pewne założenia nakładane na funkcje wagowe przez innych autorów, co ma znaczenie np. w nierównościach z wagami Czebyszewa.

Słowa kluczowe: nierówność całkowa, funkcja absolutnie ciągła

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