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(54) Title: THERMOPLASTIC ARTICLE WHICH EXHIBITS ANGULAR METAMERISM

(57) Abstract

There is described herein a new type of color effect for an article comprising a thermoplastic that is visually appealing to consumers. Specifically, there is described an article comprising a transparent thermoplastic matrix having a pigment which exhibits angular metamerism dispersed therein. The viewer looking at the article sees a viewing surface of the thermoplastic matrix. When the surface of the thermoplastic matrix opposite the viewing surface is opaque, or the matrix itself is sufficently thick as to appear opaque, the color of the viewing surface will shift color depending on the viewing angle with respect to the normal of the viewing surface.

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THERMOPLASTIC ARTICLE WHICH EXHIBITS ANGULAR METAMERISM

BACKGROUND OF THE INVENTION

This application relates generally to an article comprising a pigment which exhibits angular metamerism. The pigment is dispersed in a transparent thermoplastic matrix. This application further relates to a method for producing a thermoplastic article which exhibits angular metamerism.

Various efforts have been made to enhance the surface appearance of thermoplastic articles to make them more visually exciting to the consumer. For example, various persons have devised ways to color thermoplastics while avoiding unacceptable adverse effects on the physical properties of the thermoplastic. These techniques have improved the commercial acceptance of plastics by allowing plastic parts to be color matched to colored parts made from other materials. For example, the exterior of a car body may have both steel and plastic parts which are painted to have a matching color.

As thermoplastic parts are increasingly used for various different applications in the marketplace, it has become commercially important for manufacturers to devise new color effects for thermoplastics to make them more visually appealing to the consumer. It is also desirable to create color effects that will not be ruined by minor scratches or chips in the thermoplastic which may be caused by routine wear.

SUMMARY OF THE INVENTION

There is described herein a new type of color effect for an article comprising a thermoplastic that is visually appealing to consumers. Specifically, there is described an article comprising a transparent thermoplastic matrix having a pigment which exhibits angular metamerism (hereinafter

"angular metamerism pigment") dispersed therein. The viewer looking at the article sees a viewing surface of the thermoplastic matrix. When the surface of the thermoplastic matrix opposite the viewing surface is opaque, the color of the viewing surface will shift color depending on the viewing angle with respect to the normal of the viewing surface. It is typically necessary to have an opaque backing opposite the viewing surface because otherwise the article will simply look like a translucent gray part. The opaque backing may be a coating or a substrate. Alternatively, a part lacking an opaque backing will also give the desired visual effect provided it is sufficiently thick and has a sufficiently high loading of the angular metamerism pigment as to appear opaque.

It is also possible to obtain different attractive color effects by further incorporating an optical brightening agent, a light diffuser, additional pigments and/or a fluorescent dye in the thermoplastic matrix.

It is advantageous to incorporate the angular metamerism pigment in a thermoplastic matrix, rather than as a coating, to prevent minor chips and scratches from materially changing the appearance of the part.

It is also advantageous to incorporate a coating of a transparent thermoplastic matrix with dispersed angular metamerism pigment on a thermoplastic substrate lacking said pigment because the substrate's physical properties will not be adversely affected by the pigment. Therefore, the part, as a whole, will maintain its desirable physical characteristics.

Other features and aspects of the invention will become better understood with reference to the appended claims.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

The transparent thermoplastic matrix described above may be any transparent thermoplastic material which is compatible with angular metamerism pigments. Preferred transparent thermoplastic materials include polycarbonates, polyetherimides, transparent polyimides, transparent polyamides (nylons), polyesters, transparent polycarbonate-polyester blends, polysulfones, polyether and polyphenyl sulfones, styrene acrylonitrile (SAN), polystyrene, and miscible transparent polystyrene-polyphenylene oxide (PS-PPO) blends, acrylics, polycarbonate-polysiloxanes, polyetherimide-polysiloxanes, polyarylates, and blends and copolymers of all of the above. More preferred transparent thermoplastic matrix materials are polycarbonate homopolymer or copolymers, polyester carbonates and polyethylene terephthalate (PET). The most preferred matrix material is an aromatic polycarbonate homopolymer based primarily on the bisphenol-A monomer. The synthesis of such materials is well known in the art. For example, U.S. Patent No. 5,364,926 describes the melt process for making polycarbonate, and is incorporated by reference herein.

Angular metamerism pigments comprise a series of stacked thin film platelets of inorganic materials. Methods of making these pigments are described, for example, in U.S. Patent Nos. 5,059,245; 5,569,535 and 4,434,010, which are all hereby incorporated by reference. Angular metamerism pigments have the unique property that their color changes depending on the angle at which they are viewed. These pigments have been incorporated in security inks used in newer United States $20.00, $50.00 and $100.00 bills, in order to prevent forgery. Many inorganic pigments can not be successfully incorporated into thermoplastics because they cause unacceptable degradation

of the plastic. However, the angular metamerism pigments do not degrade polycarbonate to an unacceptable extent.

To make a thermoplastic article which exhibits angular metamerism, one typically must provide an opaque backing on the surface opposite the viewing surface. In a preferred embodiment of the invention, a film of the transparent thermoplastic matrix (e.g., polycarbonate) with dispersed angular metamerism pigment therein, is coated on an opaque thermoplastic substrate (e.g., polycarbonate containing a black pigment, or dyestuff, or a combination of dyestuffs that gives a black color). This film may be coated on the substrate by a casting method or by an extrusion method. By applying a film to a substrate, a small amount of the angular metamerism pigment may be used to produce a visual effect on a relatively large part. In contrast, it would be prohibitively expensive to use enough angular metamerism pigment to produce the same effect throughout the entire part. Alternatively, the layer of transparent thermoplastic matrix could be coated with an opaque layer, such as paint, on the side opposite the viewer. One could also make a formed thermoplastic matrix film comprising the angular metamerism dye, and injection molding an opaque plastic behind the formed film. The same effect could be achieved by printing an opaque layer on the back of the formed film, and injection mold thermoplastic (clear or opaque) behind the film. In still another embodiment of the invention, a layer of the transparent thermoplastic matrix with dispersed angular metamerism pigment is extruded on a thermoplastic substrate.

The thermoplastic matrix may optionally further contain an optical brightening agent, a light diffuser, additional pigments and/or a fluorescent dye. Adding an optical brightening agent helps produce a deeper color and an enhanced metallic appearance for the article. Suitable optical brightening agents include aromatic stilbene derivatives, aromatic benzoxazole derivatives,

or aromatic stilbene benzoxazole derivatives. Among these optical brightening agents, Uvitex OB from Ciba Specialty Chemicals [2,5-bis(5'-tert-butyl-2-benzoxazolyl)thiophene] is preferred.

Adding a fluorescent dyestuff modifies the color of the article, while retaining the angular metamerism effect. Suitable fluorescent dyestuffs include Permanent Pink R (Color Index Pigment Red 181, from Clariant Corporation), Hostasol Red 5B (Color Index #73300, CAS # 522-75-8, from Clariant Corporation) and Macrolex Fluorescent Yellow 10GN (Color Index Solvent Yellow 160:1, from Bayer Corporation). Among these, Permanent Pink R is preferred.

It may also be desirable to add a light diffuser to the transparent thermoplastic matrix. Suitable light diffusers included polytetrafluoroethylene, zinc oxide, and polymethylmethacrylate. Among these, Techpolymer MBX-series crosslinked polymethylmethacrylate microspheres, which are available in various diameters from Nagase America (e.g., 5 -50 micron avg. diameter), are preferred.

It may also be advantageous to add stabilizers and/or mold release agents to the transparent thermoplastic matrix.

Any type of pigment that is well known for inclusion in thermoplastic materials can also be added to the thermoplastic matrix. Preferred pigments include titanium dioxide, zinc sulfide, carbon black, cobalt chromate, cobalt titanate, cadmium sulfides, iron oxide, sodium aluminum sulfosilicate, sodium sulfosilicate, chrome antimony titanium rutile, nickel antimony titanium rutile, zinc oxide, and polytetrafluoroethylene.

It may also be advantageous to include various chemicals to prevent degradation of the thermoplastic matrix due to exposure to UV light (hereinafter "UV stabilizers"). Suitable UV stabilizers include substituted benzotriazoles, or triazines, or tetraalkylpiperidines. The UV stabilizers may be mixed into the thermoplastic matrix, or they can be included only in a "hardcoat" transparent protective layer which is applied over the viewing surface.

To the resin composition according to the invention may further be added other resins and additives such as reinforcing agents, fillers, impact modifiers, heat resisting agents, antioxidants, anti-weathering agents, lubricants, nucleating agents, plasticizers, flame retardants, flow-improving agents and anti-statics. These additives may be introduced in a mixing or molding process, provided the properties of the composition are not damaged.

The reinforcing fillers may be metallic fillers such as fine powder aluminum, iron, nickel, or metal oxides. Non-metallic fillers include carbon filaments, silicates such as mica, aluminum silicate or clay, talc and asbestos, titanium oxide, wollastonite, novaculite, potassium titanate, titanate whiskers, glass fillers and polymer fibers or combinations thereof. Glass fillers useful for reinforcement are not particularly limited in their types or shapes and may be, for instance, glass fibers, milled glass, glass flakes and hollow or solid glass beads. Glass fillers may be subjected to surface treatment with coupling agents such as silane or titanate-type agents to enhance adhesion with resin.

Reinforcing fillers are preferably used in an amount sufficient to yield the reinforcing effect, usually 1 to 60% by weight, preferably 5 to 50% by weight, based on the total weight of the composition. Glass fibers, or a combination of glass fibers with talc, mica or aluminum silicate are preferred

reinforcing agents. These fibers are preferably about 0.00012 to 0.00075 inches long.

In an exemplary embodiment of the invention, a polycarbonate derived from brominated bisphenol is added as a flame retardant. When such brominated polymers are added, inorganic or organic antimony compounds may further be blended in the composition to synergistically enhance flame retardance introduced by such polycarbonate. Suitable inorganic antimony compounds are antimony oxide, antimony phosphate, $KSb(OH)_6$, NH_4SbF_6 and Sb_2S_3. A wide variety of organic antimony compounds may also be used, such as antimonic esters of organic acids, cyclic alkyl antimonite esters and aryl antimonic acid compounds. Examples of typical organic antimony compounds are potassium antimony tartrate, antimony salt of caproic acid, $Sb(OCH_2CH_3)_3$, $Sb[OCH(CH_3)CH_2CH_3]_3$, antimony polymethylene glycorate and triphenyl antimony. A preferred antimony compound is antimony oxide.

Phosphites (e.g., aromatic phosphite thermal stabilizers), metal salts of phosphoric and phosphorous acid, hindered phenol antioxidants, and aromatic lactone radical scavengers may also be added as stabilizers or antioxidants.

Suitable antistatic agents include, but are not limited to, phosphonium salts, polyalkylene glycols, sulfonium salts and alkyl and aryl ammonium salts.

Suitable mold release agents include, but are not limited to, pentaerythritol tetracarboxylate, glycerol monocarboxylate, glycerol triscarboxylate, polyolefins, alkyl waxes and amides.

To prepare the resin composition of the invention, the components may be mixed by any known methods. Typically, there are two distinct mixing steps: a premixing step and a melt mixing step. In the premixing step, the dry

ingredients are mixed together. This premixing step is typically performed using a tumbler mixer or a ribbon blender. However, if desired, the premix may be manufactured using a high shear mixer such as a Henschel mixer or similar high intensity device. The premixing step must be followed by a melt mixing step where the premix is melted and mixed again as a melt. Alternatively, it is possible to skip the premixing step, and simply add the raw materials directly into the feed section of a melt mixing device via separate feed systems. In the melt mixing step, the ingredients are typically melt kneaded in a single screw or twin screw extruder, a Banbury mixer, a two roll mill, or similar device.

The articles according to present invention may then be manufactured by molding the melt mixed material into various shapes using an injection molding machine. Alternatively, a film can be formed on a substrate for example, by insert molding, in-mold decorating, laminating, co-injection or co-extrusion. For example, a film can be extruded onto sheet or another film, which can then be thermoformed, vacuum-formed, or shaped in some manner. All other known methods for creating multi-layer articles are also suitable.

The present invention is further described by way of the following examples. These examples are intended to be representative of the invention, and are not in any way intended to limit its scope.

EXAMPLES

The following experiments were performed to make various formulations according to the invention. All of the amounts given are based on 100 parts of 100 grade LEXAN® resin (bisphenol-A based polycarbonate homopolymer). The base formulation comprised the 100 parts LEXAN® resin mixed with 0.03 parts of Alkanox 240 (phosphite stabilizer) (i.e., 2,4-di-tert-

butylphenyl phosphite (3:1), CAS # 31570-04-4) purchased from Great Lakes Chemical, and 0.2 parts of a ChromaFlair angular metamerism effect pigment (Flex Products).

For each of the four ChromaFlair pigments, four separate compositions were prepared as summarized in Tables I - IV below. The ChromaFlair pigments used were silver/green 060, purple/orange 300, green/purple 190 and red/gold 000, for Tables I - IV, respectively. In each table, the first composition contained the base formulation together with 0.05 parts of Uvitex OB optical brightener; the second composition contained the base formulation together with 0.03 parts of Permanent Pink R fluorescent dye, which is available from Clariant Corporation; the third composition contained the base formulation together with 0.25 parts Techpolymer MBX-5 (i.e., microspheres of polymethylmethacrylate having an average diameter of 5 microns); and the fourth composition contained the base formulation together with 0.00465 parts carbon black pigment, 0.00460 parts of the Macrolex Blue RR dyestuff (Color Index Solvent Blue 97), and 0.00115 parts of the Macrolex Violet dyestuff (Color Index Solvent Violet 13). Both Macrolex dyestuffs were obtained from Bayer Corporation.

All of these samples were prepared by blending together the above-described ingredients and extruding them in a single or twin screw extruder with the zone temperatures set to between 480 and 550 degrees F. The pellets produced from this operation were then molded into color chips of various thickness in a Boy 15S thermoplastic molding machine set to 580 degrees F in the barrel zone, and 180 degrees F in the mold. The screw in the molding machine was operated at 200 rpm.

A composite part was then prepared by molding the thin (0.010 to 0.075 inches thick) angular metamerism pigment-containing section, inserting the molded angular metamerism part in the cavity of a mold having the same shape, but a larger cross-section, and over-molding with an opaque, black colored PC resin (e.g., Lexan 141-701, which comprises carbon black). Tables I - IV, which detail the compositions prepared by this method, are given below. All amounts are given in parts per hundred resin.

Composition 1 in each of Tables I - IV, contains an optical brightener. As noted above, adding an optical brightener helps produce a deeper color and an enhanced metallic appearance for the article. Composition 2 in each of Tables I - IV contains a fluorescent dye. As noted above, adding the fluorescent dye modifies the color of the article. Composition 3 in each of Tables I - IV comprises Techpolymer MBX-5, which is polymethylmethacrylate spheres having an average diameter of about 5 microns. Adding such particles makes the article translucent. Composition 4, in each of Tables I - IV comprises carbon black, and a blue and violet dye. This changes the color of the article.

Table I

Raw Materials	Composition of Batches 1-4				
Name	1	2	3	4	C
100 grade powder	100	100	100	100	100
Irgafos 168	0.03	0.03	0.03	0.03	0.03
Silver/Green 060	0.22	0.22	0.22	0.11	0.22
Optical Brightener	0.05				
Pink R (fluorescent dye)		0.03			
Techpolymer MBX-5			0.25		
Carbon Black (pigment)				0.00465	
Macrolex Blue RR (dye)				0.00460	
Macrolex Violet (dye)				0.00115	

C = Control

Table II

Raw Materials	Composition of Batches 1-4				
Name	1	2	3	4	C
100 grade powder	100	100	100	100	100
Irgafos 168	0.03	0.03	0.03	0.03	0.03
Purple/Orange 300	0.22	0.22	0.22	0.11	0.22
Optical Brightener	0.05				
Pink R (fluorescent dye)		0.03			
Techpolymer MBX-5			0.25		
Carbon Black (pigment)				0.00465	
Macrolex Blue RR (dye)				0.00460	
Macrolex Violet (dye)				0.00115	

C = Control

Table III

Raw Materials	Composition of Batches 1-4				
Name	1	2	3	4	C
100 grade powder	100	100	100	100	100
Irgafos 168	0.03	0.03	0.03	0.03	0.03
Green/Purple 190	0.22	0.22	0.22	0.11	0.11
Optical Brightener	0.05				
Pink R (fluorescent dye)		0.03			
Techpolymer MBX-5			0.25		
Carbon Black (pigment)				0.00465	
Macrolex Blue RR (dye)				0.00460	
Macrolex Violet (dye)				0.00115	

C = Control

Table IV

Raw Materials	Composition of Batches 1-4				
Name	1	2	3	4	C
100 grade powder	100	100	100	100	100
Irgafos 168	0.03	0.03	0.03	0.03	0.03
Red/Gold 000	0.22	0.22	0.22	0.11	0.11
Optical Brightener	0.05				
Pink R (fluorescent dye)		0.03			
Techpolymer MBX-5			0.25		
Carbon Black (pigment)				0.00465	
Macrolex Blue RR (dye)				0.00460	
Macrolex Violet (dye)				0.00115	

C = Control

The following are comments on certain representative samples described in Tables I - IV.

Table I, Batch 4

When viewed normal to the surface of the plaque (the angle between the surface and the viewing vector is 90 degrees), this sample has an intensely metallic, dark gray appearance. This color was much darker than the light silver color of a control plaque when viewed from the same angle. If this plaque is viewed from an angle 30 degrees from normal (the angle between the surface and the viewing vector is 60 degrees), the color appears dark metallic green.

Table III, Batch 1

When viewed normal to the surface, the plaque has a bright metallic green appearance, which appears more metallic than the control plaque. If this is viewed at an angle 30 degrees from normal, the color appears metallic purple.

Table III, Batch 3

This sample retained the same color as the control. However, it appeared less metallic when viewed from various angles.

Table IV, Batch 2

When viewed normal to the surface the plaque appeared slightly brighter and more red than the control plaque. If this plaque is viewed at an angle 30 degrees from normal, the color is an intense reddish copper, compared to a brown-gold color for the control plaque.

In alternate embodiments of the invention, the article can be formed by other methods. For example, the thick opaque section can be molded first and then inserted into a thicker mold cavity where the thin angular metamerism section will be over-molded. Other molding methods also exist, whereby the

part remains in the variable thickness mold cavity as the first section, and then the second, are injected into the mold from different molding machine barrels which contain the angular metamerism effect resin and the opaque resin. Another method involves partially filling the mold cavity from one molding machine barrel and the mold moving so that the remainder of the cavity can be filled from another molding machine barrel. These, and any other conventional forming methods, can be used.

It should be remembered that the above examples merely show representative compositions according to the invention. For example, one could make alternative embodiments of the invention wherein the opaque backing is LEXAN polycarbonate comprising different pigments other than carbon black. Also, one could prepare compositions comprising other Chroma Flair angular metamerism pigments, such as: gold/silver 080, cyan/purple 230, blue/red 280 and magenta/gold 330, or any angular metamerism pigments made by others. The compositions described herein may be incorporated in all varieties of articles where thermoplastics are suitable, including, but not limited to business equipment enclosures, enclosures for portable electronic items, parts for automobile bodies and other consumer articles. The article is not limited in any way with respect to shape, size or use.

WHAT IS CLAIMED IS:

1. An article which comprises an opaque backing and a transparent thermoplastic matrix affixed to the backing wherein the matrix has an angular metamerism pigment dispersed therein.

2. The article according to claim 1, wherein said matrix is selected from the group consisting of polycarbonates, polyetherimides, polyimides, polyamides, polyesters, polycarbonate-polyester blends, polysulfones, polyether and polyphenyl sulfones, styrene acrylonitrile, polystyrene, miscible polystyrene-polyphenylene oxide blends, acrylics, polycarbonate-polysiloxanes, polyetherimide-polysiloxanes, polyarylates, and blends and copolymers of all of the above.

3. The article according to claim 2, wherein the matrix is selected from the group consisting of polycarbonate homopolymer or copolymers, polyester carbonates, and polyethylene terephthalate.

4. The article according to claim 3, wherein the matrix is polycarbonate homopolymer.

5. The article according to claim 1, wherein the opaque backing is any solid material.

6. The article according to claim 5, wherein the opaque backing is a thermoplastic.

7. The article according to claim 6, wherein in the opaque backing is a polycarbonate substrate.

8. An article according to claim 1, wherein the opaque backing is a printed opaque layer.

9. An article according to claim 1, wherein the matrix further comprises an optical brightening agent.

10. An article according to claim 1, wherein the matrix further comprises a fluorescent dye.

11. An article according to claim 1, wherein the matrix further comprises an additional pigment or dye.

12. An article according to claim 1, wherein the matrix further comprises a light diffusing agent.

13. An article which comprises a transparent thermoplastic matrix material having an angular metamerism pigment dispersed therein, wherein the matrix has a first viewing side and a second side opposite the viewing side which second side has an opaque backing.

14. The article according to claim 13, wherein said matrix is selected from the group consisting of polycarbonates, polyetherimides, polyimides, polyamides, polyesters, polycarbonate-polyester blends, polysulfones, polyether and polyphenyl sulfones, styrene acrylonitrile, polystyrene, miscible polystyrene-polyphenylene oxide blends, acrylics, polycarbonate-polysiloxanes, polyetherimide-polysiloxanes, polyarylates, and blends and copolymers of all of the above.

15. The article according to claim 14, wherein the matrix is selected from the group consisting of polycarbonate, homopolymer or copolymers, polyester carbonates, and polyethylene terephthalate.

16. The article according to claim 15, wherein the matrix is polycarbonate homopolymer.

17. The article according to claim 13, wherein the opaque backing is any solid material.

18. The article according to claim 17, wherein the opaque backing is a thermoplastic.

19. The article according to claim 18, wherein in the opaque backing is a polycarbonate substrate.

20. An article according to claim 13, wherein the opaque backing is a printed opaque layer.

21. An article according to claim 13, wherein the matrix further comprises an optical brightening agent.

22. An article according to claim 13, wherein the matrix further comprises a fluorescent dye.

23. An article according to claim 13, wherein the matrix further comprises an additional pigment or dye.

24. An article according to claim 13, wherein the matrix further comprises a light diffusing agent.

25. A method for making an article which comprises:

a) mixing together a transparent thermoplastic matrix and an angular metamerism pigment, thereby forming the matrix having the angular metamerism pigment dispersed therein,

b) forming the matrix into an article having a first viewing side and a second side opposite the viewing side, and

c) applying an opaque backing to the second side.

26. The method according to claim 25, wherein the mixing step comprises a melt mixing substep.

27. The method according to claim 26, wherein the melt mixing substep is performed using a machine selected from the group consisting of an extruder, a Banbury mixer and a two roll mill.

28. The method according to claim 26, wherein the mixing step further comprises a premixing substep.

29. The method according to claim 28, wherein the premixing substep is performed using a machine selected from the group consisting of a tumbler mixer, a Henschel mixer and a ribbon blender.

30. The method according to claim 25, wherein the opaque backing is applied to the matrix by a technique selected from the group consisting of insert molding, in-mold decorating, laminating, co-extrusion, and co-injection.

INTERNATIONAL SEARCH REPORT

Inter ›nal Application No
PCT/US 99/20408

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 B32B27/20 B29C70/58

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 B32B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 496 630 A (HAWRYLKO ROMAN B ET AL) 5 March 1996 (1996-03-05) claims column 3, line 32 -column 4, line 23 column 6, line 29 - line 44 ---	1,5,6, 13,17, 18,25-30
A	US 5 605 751 A (SUZUKI FUKUJI ET AL) 25 February 1997 (1997-02-25) column 2, line 42 -column 3, line 4 column 4, line 20 -column 7, line 44; figures ---	1-30
A	US 5 192 609 A (CARROLL JR JOHN F) 9 March 1993 (1993-03-09) column 3, line 11 - line 54 column 5, line 23 - line 64 column 7, line 28 - line 48 --- -/--	1-30

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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