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(54) Process for making a carbon heat source and smoking article including the heat source and a flavor generator.

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Description

The present invention relates to a process for making a carbon source and to a smoking article comprising the carbon source and a flavor generator. More particularly, the present invention relates to a process for producing a carbon source from a preformed ligno-cellulosic material and to a smoking article, such as a cigarette, which includes the carbon source and a flavor generator.

One previously disclosed smoking article comprises a tube formed of combustible material which has a mouthpiece attached at one end. An axial inner tube of material, which is breakable when heated, is contained within the tube of combustible material and is coated on its inner surface with an additive material such as nicotine. Thus, on smoking, hot gases are drawn through the inner tube and release the nicotine in the form of an aerosol for inhalation by the smoker. With this device, however, there is an appreciable loss of nicotine and other desirable compounds, such as flavorants, during smolder. There is also a tendency for the inner tube to protrude unattractively from the burning end during smoking.

Another such cigarette-simulating smokeable device for releasing an aerosol into the mouth of a smoker comprises a rod of fuel having a longitudinally extending passage therethrough and a chamber in gaseous communication with an end of the passage whereby during smoking hot gases from the burning fuel rod enter the chamber. Inhalant material is located in the chamber which, when contacted by the hot gases during smoking, forms an aerosol for inhalation by the smoker. The chamber has, at an end remote from the fuel rod, a mouth-end closure member which is permeable to the aerosol. The chamber and the mouth-end closure member of this smoking article are of unitary construction and are formed by molding or extruding a conventional smoke filter plug to provide a chamber to contain the inhalant material. Preferably, the fuel rod is a molding or extrusion of reconstituted tobacco and/or tobacco substitute. The wall of the fuel rod is preferably impermeable to air.

A smoking article comprising a tubular heat source comprising heat-treated cellulose and a flavour generator disposed adjacent to the mouth-end of the article and comprising a substrate material containing at least one thermally releasable flavourant is described in FR-A-2469133. The heat source is a fuel rod extruded or molded from tobacco or a tobacco substitute or is formed of a mixture of tobacco substitute material and carbon or alternatively formed of other suitable combustible material, eg wood pulp, straw end heat-treated cellulose or an SCMC and carbon mixture.

A process for producing a combustible carbonised material by pyrolyzing preformed ligno-cellulosic material in a non-oxidizing atmosphere for a time of about 60 minutes is described in GB-A-1481056. This process comprises the thermal reaction of a cellulosic material in a non-oxidizing atmosphere and at a temperature within the range of 275 to 750 ° C until the weight loss of the cellulosic material is at least 60% and in which the cellulosic material subjected to the thermal reaction contains 3 to 15% by weight of an alkaline earth metal salt. The product is to be used as a filler in cigars, cigarettes and pipes and/or as a wrapper for cigars and cigarettes.

The present invention provides a process for producing a carbon heat source which is substantially tasteless when fabricated as a smoking article and smoked. This process is characterized by pyrolizing a preformed article in a continuously exchanged inert atmosphere at a temperature in the range of 800 ° to 1100 ° C for 0.5 to 3 hours, cooling the pyrolized article in the inert atmosphere at a rate of 500 ° to 10 ° C per hour to a temperature within the range of 275 ° to 25 ° C, and then subjecting the pyrolized article to at least one additional treatment selected from oxygen absorption, water desorption, and salt impregnation with subsequent heat treatment to obtain a tasteless carbon heat source.

The present invention also relates to a smoking article of the type described above, characterized in that the heat source is a carbon heat source produced by the process of the present invention having a porosity sufficient to support combustion and a density such that puff-induced air flow passes through the tube.

As exemplified by its preferred embodiments herein, the process of the present invention comprises three basic steps: a pyrolysis step, a controlled cooling step, and at least one additional process step selected from an oxygen absorption step, a water desorption step, and a salt impregnation and subsequent heat treatment step.

The pyrolysis step is carried out in an inert atmosphere in order to avoid combustion of the preformed article. Typically, the preformed ligno-celluslosic article is pyrolyzed in an oven which has controlled temperature zones and a quartz reaction chamber in which the articles to be pyrolyzed are placed. The quartz chamber is connected to a source of an inert gas, such as dry nitrogen or argon, and purged in order to remove the air. Throughout the process, a continuous flow of inert gas is passed through the quartz reaction chamber, hereinafter referred to as the pyrolyzing chamber, so that the inert atmosphere is continuously exchanged, whereby the volatiles driven off during pyrolysis are removed from the pyrolyzing

chamber. This continuous exchange is believed to be important to the production of an essentially tasteless carbon heat source.

The article to be pyrolyzed is heated to a temperature within the range of from about 800° to about 1100° C, and more preferably from about 950° to about 1000° C, and is maintained at this temperature for from about 0.5 to about 3 hours, preferably from about 0.5 to about 1.5 hours, and more preferably from about 0.75 to about 1.25 hours. Typically, the inert gas employed is dry nitrogen and the flow rate through the pyrolyzing chamber is adjusted to within the range of from about 0.5 to about 5 liters per minute, preferably from about 1 to about 1.5 liters per minute, during pyrolysis. During pyrolysis, the ligno-cellulosic material generally experiences a weight loss of about 70% to about 80% and a dimensional shrinkage generally within the range of about 30% to about 35%.

Upon completion of pyrolysis, the pyrolyzed material is gradually cooled to a temperature within the range of from about 275° C to about 25° C, preferably about 100° C to about 25° C. Typical rate of cooling will be from about 500° to about 10° C per hour, preferably from about 100° to about 60° C per hour. It is important that the rate of cooling be gradual and controlled. It has been observed that a rapid quench, such as immersion in liquid nitrogen, will adversely affect the burn properties of the pyrolyzed material.

According to the oxygen absorption step, which functions to add oxygen to the pyrolyzed article, air or oxygen is gradually introduced into the inert gas stream as the temperature falls to within the range of from about 275° C to about 25° C, preferably from about 100° C to about 35° C. While oxygen absorption may be initiated at temperatures as high as 530° C or as low as 25° C, it is preferred to operate within the above ranges. The oxygen is gradually introduced and the flow rate increased until the oxygen substantially replaces the inert gas. It is important to gradually introduce the oxygen as the cooling continues in order to avoid excessive oxidation of the pyrolyzed material. Preferably, the oxygen is introduced such that the ratio of the volume of nitrogen to the volume of oxygen is within the range of about 1:4 to about 8:1, most preferably about 4:1. During the oxygen absorption step, the pyrolyzed material is either at or is cooled to room temperature.

According to the impregnation and heat treatment step, the pyrolyzed article, which has been cooled to room temperature either with or without the oxygen absorption step, is first impregnated with an aqueous solution of salts of a cation selected from the group consisting of K^{+}, Fe^{+2}, Fe^{+3}, Mg^{+2}, Mn^{+2}, Ca^{+2} and mixtures thereof. The pyrolyzed material is impregnated such that it contains from about 0.5 to about 11% of the cation on a dry weight basis, preferably from about 1% to about 3%. Any means known to those skilled in the art may be used to impregnate the pyrolyzed material with the salt solution. One particularly preferred means is vacuum impregnation. After impregnation, the material is then dried at a temperature within the range of from about 40° to about 100° C, preferably from about 50° to about 70° C, in vacuum.

The dried, impregnated, pyrolyzed material is then gradually heated to a temperature within the range of from about 550° to about 750° C, preferably to about 650° C, in an inert atmosphere and is maintained at this temperature for from about 5 to about 60 minutes, preferably from about 15 to about 30 minutes. The material is then cooled in the inert atmosphere.

According to the water desorption step, which, when employed, is preferably the final process step, the pyrolyzed article is subjected to a desiccant environment for at least about 8 hours preferably from about 12 hours to about 48 hours. The purpose of this step is to maintain an existing, or establish and maintain, a relatively moisture-free state in the carbon heat source. This step is preferably practiced by placing the pyrolyzed article in a desiccator containing $CaSO_4$. It has been observed that this process step improves the burn properties of the carbon heat source.

Any one or combination of the additional process steps may be employed. When salt impregnation and oxygen absorption are both employed, it is preferred that the oxygen absorption step follow the impregnation step.

As the ligno-cellulosic material, tobacco, peanut shells, coffee bean shells, paper, cardboard, bamboo, oak leaves, or a similar such material is suitably employed. The material may preferably be admixed with a binder, such as hydroxypropyl cellulose prior to formation into the desired shape.

The ligno-cellulosic material is preformed, prior to pyrolysis, into the shape desired upon completion of the pyrolysis and subsequent treatment steps, taking into account the dimensional shrinkage experienced during pyrolysis. Extrusion, rolling, injection-molding or the like may be employed to shape the article. preferably, extruded, substantially tube-shaped articles with porous material located in the core of the tubes are employed. The article, once pyrolyzed, must be sufficiently rigid to maintain the shape of the smoking article during smoking and must have a porosity sufficient to absorb the salt solution and oxygen, when employed, yet less porous than the material in the core, when present, so that the gaseous combustion products will flow through the central passage to the flavor source and not through the pyrolyzed material.

The present invention also relates to smoking articles comprising a flavor generator and a carbon heat

source. The carbon heat source is the pyrolyzed material prepared according to the process of the present invention. While the carbon source may be prepared in any of the various commercially available shapes of smoking articles, the smoking article will be described with respect to a cigarette.

According to this embodiment, the smoking article is prepared by pyrolyzing a tube-shaped article of lignocellulosic material and then attaching the flavor generator adjacent the mouth end thereof. The tube-shaped carbon heat source may be formed with a porous, preferably open-cell foam, combustible material in the core, as by a co-extrusion process, or, preferably, with at least one porous, combustible plug disposed within the passage. When only one plug is employed, it is preferably disposed at the coal end of the cigarette to prevent flash jetting while the cigarette is being lit. When a porous core is employed, the core material is less dense than the surrounding tube-shaped material so that the combustion gases will flow through the central core to the flavor generator rather than through the carbon source. By selecting the type and amount of material placed in the passage, the temperature of the gases reaching the flavor generator can be established within a range such that thermally releasable flavorants are released without undergoing thermally induced decomposition to products which are not desirable as flavorants.

The flavor generator comprises a substrate material, such as alumina, magnesium hydroxide, zeolites, glass wool, charcoal, tobacco filler, fuller's earth, natural clays, and activated clays, which is impregnated with at least one thermally releasable flavorant, or which inherently contains at least one thermally releasable flavorant. The flavoring agent may consist of any suitable blend of natural or synthetic flavorants such as nicotine, glycerol, menthol, vanilla, eucalyptol, octyl acetate, orange, mint, or isoamyl isovalerate. The flavor generator is preferably cylindrical and of a diameter substantially equal to the diameter of the carbon source, and may be placed in abutting end-to-end relation to the carbon source or may be spaced therefrom. The carbon source and flavor generator may be wrapped in cigarette paper and, if desired, a conventional filter, such as cellulose acetate filter, may be placed after the flavor generator and joined thereto by tipping paper or the like. The flavor generator may comprise a flavored, foamed core containing readily volatilized flavors that are not subject to thermal degradation.

As the hot gases flow through the channel or bore in the carbon source and over the flavor generator, most of the flavors are distilled from the substrate material and the distillate is carried toward the smoker's mouth due to the drawing action. As the flavor-laden gases pass away from the flavor generator toward the cooler portion of the cigarette, the oils contained in the distillate recondense into relatively small droplets, forming a mist or aerosol, and pass into the mouth and nose of the smoker where they create a sensation by taste and smell. A sufficient amount of flavorant should be provided such that the flavorant is continuously released until the smoking article is extinguished.

When extruded tobacco articles are employed as the ligno-cellulosic material in the present process, they are preferably prepared according to the process disclosed in US-A 4 347 855 (see also GB-A 2 078 087 or DE-A 31 18 472.

Examples

The following examples present illustrative but non-limiting embodiments of the present invention. A comparative example is also presented.

In each of the following examples 1 through 9, extruded tobacco tubes prepared according to the method disclosed in U.S. Patent 4,347,855 were employed as the preformed ligno-cellulosic material and were pyrolyzed in a Lindberg, 3-zone furnace having a chamber 152 mm in diameter and 914 mm long surrounding a quartz tube pyrolizing chamber 134 mm in diameter and 1.32m long. The furnace was equipped with seven thermocouples spaced along the length of the quartz tube and could achieve a maximum temperature of about 1200 ° C.

Example 1

Extruded tobacco tubes were prepared using -20 + 30 mesh (0.60-0.84 mm) tobacco. Two sets of tobacco tubes were employed; one set had an outside diameter of 8 mm and an inside diameter of 5 mm, and the other had an outside diameter of 12 mm and an inside diameter of 5 mm. The tobacco tubes were pyrolyzed according to the procedure summarized below in Table 1.

Table 1

Elapsed Time (minutes)	Thermocouple Readings (°C) 1	2	3	4	5	6	7	Comments
0								Tobacco tubes placed in quartz chamber and chamber purged with N_2 at a flow rate of 1 l/min. Furnace
90	22	22	21	21	21	21	22	turned on.
97	52	97	94	78	94	95	59	
179	552	757	837	850	789	692	517	
190	597	803	880	891	829	733	573	
227	711	903	966	972	912	825	657	
258	752	917	967	972	917	840	684	
280	769	922	967	966	919	844	694	
285	772	924	969	967	920	846	697	Furnace turned off.
308	741	839	862	855	813	762	646	
321	712	796	815	806	767	721	613	
340	670	745	760	749	711	671	570	
350	649	721	735	723	687	648	550	
360	631	700	712	700	664	628	532	
370	612	679	691	678	643	607	514	
1347	103	120	123	114	105	31	99	
1354								Furnace lid lifted.
1361	82	91	88	86	76	28	80	
1507	27	29	28	26	25	20	25	
1815	20	21	21	20	20	20	20	
1816								Gas flow changed from 1.05 l/min. of N_2 to 1.76 l/min. of air and N_2. The air/N_2 ratio was 700/1050
1821	20	20	21	20	20	19	20	
1826	20	20	21	20	20	19	20	N_2 turned off; air intro-
1831	20	20	21	20	20	19	20	duced at a flow rate of
1846	20	21	21	21	20	20	20	0.75 l/min.
1851	20	21	21	21	21	20	21	
1861	20	21	21	21	21	21	21	Air flow turned off.
1876	20	21	22	21	21	21	21	
2763	21	21	21	21	21	21	21	
2776								Pyrolyzed tobacco tubes removed from quartz chamber.

The pyrolyzed samples were measured and weighed and it was determined that the samples experienced an average weight loss of 84.7%, an average decrease in length of 33.66%, an average decrease in outside diameter of 33.25%, and an average decrease in inside diameter of 33.05%. The pyrolyzed samples burned statically when lit. Static burning occurs when a cigarette rod continues to smoulder, once it has been lit, in the absence of air drafts and puff induced air flow.

Example 2

Two sets of extruded tobacco tubes were pyrolyzed; one set had an outside diameter of 12 mm and an inside diameter of 5 mm, the other set had an outside diameter of 8 mm and an inside diameter of 2.5 mm. The tobacco tubes were pyrolyzed according to the procedure summarized below in Table 2.

Table 2

Elapsed Time (minutes)	Thermocouple Readings (°C) 1	2	3	4	5	6	7	Comments
0								Tobacco tubes placed in quartz chamber; N_2 purge initiated at 1.05 l/min. flow rate. Furnace turned on.
185								
187	24	25	25	25	26	26	26	
207	178	269	325	258	265	259	192	
279	546	670	762	759	680	607	468	
290	562	678	763	758	679	609	477	
317	589	691	765	755	677	614	487	
324	595	694	765	755	677	614	490	
349	609	700	769	752	675	615	494	
462	642	718	769	750	672	619	507	
465								Furnace turned off.
483	619	668	696	675	603	564	491	
500	591	630	650	626	558	526	446	
1445	103	98	99	90	83	84	80	N_2 flow rate increased to 4.2 l/min.
1446								Furnace lid lifted.
1467	62	59	58	54	47	47	46	
1494	44	45	46	42	41	37	37	N_2 flow rate reduced to 1 l/min.
1564	32	35	36	34	31	31	30	
1953								Air introduced at a flow rate of 1 l/min.; flow rate of air plus flow rate of N_2 = 2.05 l/min.
1955	24	25	25	27	25	25	25	
1960	24	25	26	28	26	26	26	
1965	24	25	25	26	25	25	25	
2916	22	22	23	23	23	23	23	
3066								Air flow rate increased to 4 l/min; flow rate of air plus flow rate of N_2 = 5 l/min.
3067	23	23	23	23	24	24	24	
3243	23	23	23	23	24	24	24	
3245								N_2 flow and air flow turned off; samples removed from quartz chamber.

The pyrolyzed tobacco tubes evidenced a 72% weight loss and a 4 to 4.5% dimensional decrease for the larger diameter tubes and a 69% weight loss and 37.5% dimensional decrease for the smaller diameter tubes.

Example 3

Extruded tobacco tubes were pyrolyzed according to the procedure summarized below in Table 3.

TABLE 3

Elapsed Time (minutes)	Thermocouple Readings (°C) 1	2	3	4	5	6	7	Comments
0								Tobacco tubes placed in quartz chamber; N_2 purge initiated at an N_2 flow rate of 1.05 l/min.
1440								
1441	17	18	19	18	18	18	18	Furnace turned on.
1448	37	85	84	65	74	52	--	
1464	186	331	377	336	314	199	209	
1471	233	402	459	432	399	162	256	
1476	260	442	506	485	447	393	287	
1486	323	523	595	585	537	468	337	
1525	510	730	811	813	759	661	498	
1744	684	833	869	860	806	743	608	
1745								Furnace turned off.
1751	678	811	839	829	771	718	600	
2079								N_2 flow rate increased to 2.3 l/min.
2889	94	92	93	84	77	77	75	N_2 flow rate increased to 2.6 l/min.
2936	86	88	88	82	77	77	72	Furnace lid lifted.
3035	36	33	34	32	30	29	29	
3170	28	27	27	26	25	25	25	
3173								Air introduced at a flow rate of 1.05 l/min.; N_2 flow rate reduced to 1.05 l/min.
3175	28	27	27	26	25	24	24	
3184	27	27	27	26	25	24	24	
3189								Air flow rate increased to 2 l/min.
3192	27	26	27	26	25	24	24	
3198								Air flow rate increased to 3 l/min.
3199	27	26	26	25	25	24	24	
3211	27	26	26	25	25	25	24	
3212								Air flow rate increased to 4 l/min.
3215	26	26	26	25	25	24	24	
3220								N_2 turned off.
3227	26	25	26	25	25	25	25	
3233	26	25	26	25	25	24	24	
3282	25	25	25	25	24	24	24	
3291								Pyrolyzed tobacco tubes removed from quartz chamber.

The pyrolyzed tobacco tubes maintained a static burn when lit both before and after being placed in a desiccator containing $CaSO_4$ for about 48 hours. It was determined that the pyrolyzed tubes experienced a decrease in length of 27.24%, a decrease in outside diamter of 7.5%, and a decrease in inside diameter of 19.29%.

Example 4

Two sets of extruded tobacco tubes were prepared; one set from tobacco material 60% of which was below 60 mesh (0.25mm)and 40% of - 20 + 30 mesh, (0.42-0.60mm) and the other set from tobacco material 60% of which was below 60 mesh and 40% of -30 + 40 mesh. The tobacco tubes were 65 mm in length, and had an outside diameter of 8 mm and an inside diameter of 5 mm. The tobacco tubes were

pyrolyzed according to the procedure summarized below in Table 4.

Table 4

Elapsed Time (minutes)	Thermocouple Readings (°C) 1	2	3	4	5	6	7	Comments
0								Tobacco tubes placed in quartz chamber; N_2 introduced at flow rate of 9 l/min. Furnace turned on.
95								
117	136	295	331	314	316	282	217	
147	247	509	595	607	573	492	368	
240	211	316	349	359	339	311	280	
318	459	724	820	851	803	722	572	
420	524	750	828	855	819	751	621	
437	526	749	826	853	818	751	622	Furnace turned off.
1381	52	67	70	70	67	67	66	
1443	48	62	64	64	62	62	61	
1506	45	56	58	59	57	57	56	Furnace lid lifted.
1528	34	37	39	42	39	38	39	
1670	24	26	27	28	27	27	27	
1684	24	26	27	27	27	27	27	
1685								Air introduced at a flow rate of 1 l/min.
1696	24	26	27	27	26	26	26	
1832	24	26	27	27	26	26	26	
1887	24	24	25	25	25	25	25	
2850								Pyrolyzed tobacco tubes removed from quartz chamber.

Both sets of pyrolyzed tobacco tubes maintained a static burn.

Example 5

Two sets of extruded tobacco tubes were prepared; one set from tobacco material 60% of which was -60 mesh and 40% was -30 + 40 mesh, and the other set from tobacco material 60% of which was -60 mesh and 40% was -20 + 30 mesh. The tobacco tubes had an outside diameter of 12 mm and an inside diameter of 7 mm. The tobacco tubes were pyrolyzed according to the procedure summarized below in Table 5.

Table 5

Elapsed Time (minutes)	Thermocouple Readings (°C) 1	2	3	4	5	6	7	Comments
0								Tobacco tubes placed in quartz chamber; N_2 introduced at flow rate of
7200	21	21	21	21	22	22	21	1 l/min. Furnace turned on.
7213	97	177	175	134	164	158	98	
7216	128	221	234	183	219	200	129	
7221	185	301	335	303	306	264	190	
7246	338	503	580	579	544	456	328	
7379	794	919	971	965	912	828	655	
7416	816	929	973	966	915	833	661	
7476	835	937	975	965	915	839	672	Furnace turned off.
7581	634	672	678	658	620	583	478	
7650	549	587	585	564	531	499	410	
8709	93	96	97	92	90	87	78	
8836	78	80	81	77	75	73	66	
8862	75	77	78	74	72	70	64	
8910	70	72	72	69	67	66	60	Furnace lid lifted.
8966	37	35	36	34	32	31	31	
9046								Air introduced at a flow rate of 4 l/min.; N_2 flow turned off.
9048	29	29	29	27	26	26	25	
9079	28	27	28	26	25	26	25	Samples removed from quartz chamber.

Both sets of pyrolyzed tobacco tubes maintained a static burn.

Example 6

Extruded tobacco tubes were pyrolyzed according to the procedure summarized below in Table 6.

Table 6

Elapsed Time (minutes)	Thermocouple Readings (°C) 1	2	3	4	5	6	7	Comments
0								Tobacco tubes placed in quartz chamber; N_2 introduced at a flow rate of 1 l/min. Furnace turned on.
1335								
1343	44	66	54	60	64	62	22	
1348	128	169	133	154	166	149	32	
1355	211	295	264	277	272	221	50	
1363	288	403	407	395	366	285	73	
1372	356	490	508	488	443	336	95	
1389	469	626	657	632	566	430	147	
1408	571	729	764	738	662	509	202	
1422	639	793	828	801	722	567	245	
1434	687	836	870	843	764	609	277	
1452	759	897	929	902	824	673	324	
1497	869	961	981	954	887	764	401	
1561	894	970	983	954	891	780	411	Furnace turned off.
1642	650	665	661	631	596	536	256	
1664	617	631	626	596	562	505	236	
1702	569	581	575	545	514	461	209	
1721	549	560	553	523	493	442	198	
1790	482	491	482	454	428	385	166	
2743	95	94	92	87	85	79	40	Furnace lid lifted.
2812	40	39	37	35	33	31	25	
2840	36	36	34	32	30	29	24	
2861	35	34	32	31	29	28	24	
2899	31	32	31	30	28	28	25	
2903								Air introduced at a flow rate of 4 l/min.
2905			34*					Air flow turned off.
2959	29	29	29	28	27	26	24	
2965								Air introduced at a flow rate of 4 l/min.
2970								N_2 flow turned off.
3091	26	26	26	26	25	25	23	
3206	25	25	25	25	24	24	22	Samples removed from quartz chamber.

* When TC #3 indicated a temperature rise, the air was turned off.

The samples were removed from the furnace and placed in a desiccator containing $CaSO_4$. The pyrolyzed tobacco tubes maintained a static burn.

Example 7

Four sets of extruded tobacco tubes were prepared; one set from -30 + 40 mesh tobacco particles, a second set from -20 mesh tobacco particles, a third set from -20 + 30 mesh tobacco particles, and a fourth set from -20 + 30 mesh, recycled tobacco particles. The extruded tobacco tubes were pyrolyzed according to the procedure summarized below in Table 7.

Table 7

Elapsed Time (minutes)	Thermocouple Readings (°C) 1	2	3	4	5	6	7	Comments
0								Tobacco tubes placed in the quartz chamber; N_2 introduced at a flow rate of 1 l/min. Furnace turned on.
1280								
1281	23	25	24	25	25	25	21	
1290	121	149	119	134	141	130	25	
1300	271	336	324	324	301	244	48	
1311	378	473	479	462	417	323	82	
1322	454	567	584	562	501	382	112	
1348	584	716	744	717	639	495	175	
1423	841	951	968	939	874	754	362	
1447	896	1006	1019	989	928	811	397	
1457	882	954	965	934	883	791	404	
1467	899	985	996	964	910	809	402	
1485	890	972	979	949	900	819	402	
1487								Furnace turned off.
1495	874	929	936	905	862	781	401	
1504	841	884	887	858	820	748	384	
1514	807	841	842	813	779	714	363	
1633	583	598	594	567	544	498	228	
1724	488	500	495	469	450	412	181	
1751	464	476	469	444	427	391	170	
1770	451	462	456	431	414	379	164	
2712	95	96	94	90	89	82	40	Furnace lid lifted; N_2 flow rate increased to 3 l/min.
2725	70	67	71	63	59	55	38	
2804	36	37	35	33	31	30	25	
2879	31	31	30	29	28	27	24	
2882								N_2 flow rate adjusted to 1 l/min.; air introduced at flow rate of 4 l/min.
2885	31	31	31	28	27	27	24	
2917	30	30	29	27	26	26	24	
2937	29	29	28	27	26	26	24	
3042	27	27	26	26	25	25	24	N_2 flow turned off.
3182	25	25	25	25	24	25	24	
4187	22	22	23	22	22	22	22	Samples removed from quartz chamber.

It was determined that the pyrolyzed tobacco tubes experienced a weight loss in the range of 78% to 79%, and a dimensional decrease within the range of from about 27% to about 33%. All of the pyrolyzed tobacco tubes maintained a static burn.

Example 8

Previously pyrolyzed tobacco tubes were vacuum impregnated with a saturated solution of either KNO_3, $Mg(CH_3COO)_2$, $FeCl_3$, $K_3C_6H_5O_7$, $FeCl_2$ or $MgCl_2$. The impregnated pyrolyzed tubes were dried in an oven in vacuum at 50° C, and then heat treated in the Lindberg furnace described above according to the procedure summarized below in Table 8.

Table 8

Elapsed Time (minutes)	Thermocouple Readings (°C) 1	2	3	4	5	6	7	Comments
0								Pyrolyzed tobacco tubes placed in quartz chamber; N_2 introduced at a flow rate of 1 l/min.
140	21	22	24	25	25	23	21	Furnace turned on.
146	74	71	93	91	102	48	24	
164	308	381	422	401	371	101	71	
176	403	495	545	521	464	119	116	
282	451	512	559	528	476	401	173	
331	564	624	665	638	574	490	242	
332								Furnace turned off.
416	434	453	465	440	406	366	173	
428	421	438	448	424	392	354	166	
1374	88	88	85	82	79	74	38	Furnace lid lifted.
1414	43	46	43	38	36	35	29	
1477	33	35	32	30	28	28	25	
1482								Air introduced at a flow rate of 4 l/min.
1483	33	34	32	30	28	28	25	
1484								N_2 flow turned off.
1488	33	34	34	30	28	28	25	
1496	32	33	32	30	28	27	25	
1498								Air flow rate decreased to 2 l/min.
1514	31	32	30	29	27	27	25	
1558	29	30	28	27	26	26	24	
1634	27	28	27	26	25	25	24	Air flow rate decreased to 1 l/min.
1750	25	25	25	25	24	24	23	Air flow turned off.
1835								Pyrolyzed tubes removed from quartz chamber.

The salt treated, pyrolyzed tubes containing absorbed oxygen, maintained a static burn when ignited.

Example 9

Extruded tobacco tubes were prepared from tobacco material of mesh size +60. The extruded tobacco tubes had an outside diameter of 12mm, and an inside diameter of 5mm and were pyrolyzed according to the procedure summarized below in Table 9.

Table 9

Elapsed Time (minutes)	Thermocouple Readings (°C) 1	2	3	4	5	6	7	Comments
								Tobacco tubes placed in quartz chamber and chamber purged overnight in N_2 at a flow rate of 1 l/min.
0								Furnace turned on
1	23	24	24	24	24	24	24	
19	122	226	309	241	246	249	186	
31	215	343	456	499	410	365	280	
48	303	461	600	611	559	486	369	
57	347	516	664	681	625	544	415	
101	546	724	878	897	832	740	590	
161	733	870	973	979	909	839	711	
194	759	888	975	977	910	843	723	
229	775	900	977	977	907	846	731	Furnace turned off
300	630	708	722	712	655	624	557	
399	462	561	570	556	507	484	433	
448	412	509	518	503	457	437	393	
466	395	492	500	485	440	421	379	
1427	74	98	97	92	83	83	80	Furnace lid raised
1560	33	34	34	34	30	30	30	Air flow introduced at a rate of 4 l/min.
1564	32	33	34	36	31	31	31	Air flow turned off
1590	31	32	33	32	29	29	29	Air flow turned on at a rate of 4 l/min.
1599	31	31	32	31	29	29	29	
1652	29	29	29	29	27	27	27	
1770	26	26	27	26	25	25	25	
1829	25	25	26	26	25	25	25	N_2 turned off
1886	25	26	27	26	24	24	24	
2874	22	22	22	22	21	21	21	Air flow turned off
2885								Pyrolyzed tobacco tubes removed from quartz chamber

The pyrolyzed samples were measured and weighed and it was determined that the samples experienced an average weight loss of 73.47%, and an average shrinkage loss of 31.41%. The samples would not sustain static burning.

The following example is comparative.

Comparative Example 1

Extruded tobacco tubes were prepared from tobacco material of mesh size -20. The extruded tobacco tubes, which were 90mm in length, with an outside diameter of 12mm and an inside diameter of 10mm, were pyrolyzed inside a quartz tube in the chamber of a Lindberg 55035-A oven. The oven was equipped with one thermocouple positioned over the center of the longitudinal axis of the tube. The procedure used is summarized below in Table 10.

Table 10

Elapsed Time (Minutes)	Thermocouple Reading (°C)	Comments
		Tobacco tubes placed in quartz chamber and chamber purged with N_2 at a flow rate of 1.05 l/min overnight.
0		Furnace turned on
22	725	
118	920	
148	940	
162	950	
178	960	
196	960	Furnace turned off
205	960	
215	800	
220	740	
250	510	
265	440	
290	390	
313	390	
661	390	Pyrolyzed tobacco tubes removed from quartz chamber.

The pyrolyzed samples were removed from the chamber and quenched in liquid nitrogen. The samples were then weighed and measured, and it was determined that the samples experienced an average decrease in length of 31.6%, an average decrease in outside diameter of 28.29%, and an average decrease in inside diameter of 34%. The pyrolyzed samples would not sustain static burning.

Claims

1. A process for producing a combustible carbonized material by pyrolizing preformed ligno-cellulosic material in a non-oxidizing atmosphere,the process comprising pyrolizing a preformed article in a continuously exchanged inert atmosphere at a temperature in the rang of 800° to 1100° C for 0.5 to 3 hours, cooling the pyrolized article in the inert atmosphere at a rate of 500° to 10° C per hour to a temperature within the range of 275° C to 25° C, and then subjecting the pyrolized article to at least one additional treatment selected from oxygen absorption, water desorption, and salt impregnation with subsequent heat treatment to obtain a tasteless carbon heat source.

2. A process according to claim 1, characterized by adding oxygen to the pyrolized article and then, as a final step, subjecting the pyrolized article to a desiccant environment.

3. A process according to claim 1, characterized by contacting the pyrolized article with a salt solution comprising a salt of at least one of the cations K^+, Fe^{+3}, Fe^{+2}, Mg^{+2}, Ca^{+2}, drying the article at a temperature within the range of 50° to 70° C in vacuum, gradually heating the article to a temperature of about 650° C in an inert atmosphere and maintaining the article at said temperature for 5 to 60 minutes, and then cooling the article in the inert atmosphere at a rate of 500° to 10° C per hour to a temperature within the range of 275° C to 25° C.

4. A process according to claim 3, characterized by adding oxygen to the pyrolized article after the second cooling step.

5. A process according to claim 3 or 4, characterized by subjecting the pyrolized article to a desiccant environment, as a final step.

6. A process according to any of claims 3 to 5, characterized in that the pyrolized material is contacted

with the salt solution by vacuum impregnation.

7. A process according to any of claims 1 to 6, characterized in that cellulosic material is selected from cardboard, paper, bamboo, oak leaves and extruded tobacco.

8. A smoking article comprising a substantially tubular heat source comprising heat-treated cellulose and a flavour generator disposed adjacent the mouth end of the article and comprising a substrate material containing at least one thermally releasable flavorant, characterized in that the heat source is a carbon heat source produced by a process according to any of claims 1 to 7, having a porosity sufficient to support combustion and a density such that puff-induced air flow passes through the tube.

9. A smoking article according to claim 8, characterized in that the substrate is selected from alumina, tobacco filler, magnesium hydroxide, zeolites, glass wool, charcoal, fuller's earth, natural clays, and activated clays.

10. A smoking article according to claim 8 or 9 characterized by a porous, combustible material disposed within the tube passage and having a porosity greater than the porosity of the carbon heat source.

Revendications

1. Procédé de production d'une matière carbonisée combustible par pyrolyse d'une matière lignocellulosique préformée dans une atmosphère non oxydante, le procédé consistant à pyrolyser un article préformé dans une atmosphère inerte constamment renouvelée à une température située dans l'intervalle de 800° à 1100°C pendant 0,5 à 3 heures, refroidir l'article pyrolysé dans l'atmosphère inerte à une vitesse de 500° à 10°c par heure jusqu'à une température située dans l'intervalle de 275°C à 25°C, puis soumettre l'article pyrolysé à au moins un traitement supplémentaire choisi parmi une absorption d'oxygène, une désorption d'eau et une imprégnation de sel avec traitement thermique subséquent pour obtenir une source de chaleur carbonée insipide.

2. Procédé selon la revendication 1, caractérisé en ce qu'on ajoute de l'oxygène à l'article pyrolysé puis, à titre d'étape finale, on soumet l'article pyrolysé à un milieu desséchant.

3. Procédé selon la revendication 1, caractérisé en ce qu'on met en contact l'article pyrolysé avec une solution de sel comprenant un sel d'au moins l'un des cations K^+, Fe^{3+}, Fe^{2+}, Mg^{2+}, Mn^{2+}, Ca^{2+}, on sèche l'article sous vide à une température située dans l'intervalle de 50° à 70°C, on chauffe progressivement l'article à une température d'environ 650°C dans une atmosphère inerte et on maintient l'article à cette température pendant 5 à 60 minutes, puis on refroidit l'article dans l'atmosphère inerte à une vitesse de 500° à 10°C par heure jusqu'à une température située dans l'intervalle de 275°C à 25°C.

4. Procédé selon la revendication 3, caractérisé en ce qu'on ajoute de l'oxygène à l'article pyrolysé après la seconde étape de refroidissement.

5. Procédé selon la revendication 3 ou 4, caractérisé en ce qu'à titre d'étape finale, on soumet l'article pyrolysé à un milieu desséchant.

6. Procédé selon l'une quelconque des revendications 3 à 5, caractérisé en ce que la matière pyrolysée est mise en contact avec la solution de sel par imprégnation sous vide.

7. Procédé selon l'une quelconque des revendications 1 à 6, caractérisé en ce que la matière cellulosique est choisie parmi le carton, le papier, le bambou, les feuilles de chêne et le tabac extrudé.

8. Article à fumer comprenant une source de chaleur sensiblement tubulaire comprenant de la cellulose traitée thermiquement et un générateur d'arôme disposé près de l'extrémité du côté bouche de l'article et comprenant une matière de substrat contenant au moins un agent aromatisant libérable par la chaleur, caractérisé en ce que la source de chaleur est une source de chaleur carbonée produite par un procédé selon l'une quelconque des revendications 1 à 7, ayant une porosité suffisante pour

entretenir la combustion et une densité telle qu'un courant d'air provoqué par aspiration traverse le tube.

9. Article à fumer selon la revendication 8, caractérisé en ce que le substrat est choisi parmi l'alumine, une charge pour tabac, l'hydroxyde de magnésium, les zéolites, la laine de verre, le charbon de bois, la terre à foulon, les argiles naturelles et les argiles activées.

10. Article à fumer selon la revendication 8 ou 9, caractérisé par une matière combustible poreuse disposée dans le passage du tube et ayant une porosité supérieure à la porosité de la source de chaleur carbonée.

Ansprüche

1. Verfahren zur Herstellung eines brennbaren carbonisierten Materials durch Pyrolisieren vorgeformten Ligno-Cellulosematerials in einer nicht-oxidierenden Atmosphäre, welches Verfahren das Pyrolisieren eines vorgeformten Gegenstandes in einer kontinuierlich ausgetauschten inerten Atmosphäre bei einer Temperatur im Bereich von 800° bis 1100° C über 0,5 bis 3 Stunden, das Abkühlen des pyrolisierten Gegenstands in der inerten Atmosphäre mit einer Rate von 500° bis 10° C pro Stunde auf eine Temperatur im Bereich von 275° C bis 25° C und danach das Unterwerfen des pyrolisierten Gegenstandes wenigstens einer weiteren Behandlung, ausgewählt aus Sauerstoffabsorption, Wasserdesorption und Salzimprägnierung mit anschließender Hitzebehandlung, umfaßt, um eine geschmacklose Kohlenstoffhitzequelle zu erhalten.

2. Verfahren nach Anspruch 1, gekennzeichnet durch die Zugabe von Sauerstoff zu dem pyrolisierten Gegenstand und danach, als letztem Schritt, Unterwerfen des pyrolisierten Gegenstands einer trocknenden Umgebung.

3. Verfahren nach Anspruch 1, gekennzeichnet durch das Inberührungbringen des pyrolisierten Gegenstands mit einer Salzlösung, die ein Salz wenigstens eines der Kationen K^+, Fe^{+3}, Fe^{+2}, Mg^{+2}, Mn^{+2}, Ca^{+2} umfaßt, das Trocknen des Gegenstands bei einer Temperatur im Bereich von 50° C bis 70° C im Vakuum, das allmähliche Erhitzen des Gegenstands auf eine Temperatur von etwa 650° C in einer inerten Atmosphäre und das Halten des Gegenstands bei dieser Temperatur über 5 bis 60 Minuten sowie danach das Abkühlen des Gegenstands in der inerten Atmosphäre mit einer Rate von 500° bis 10° c pro Stunde auf eine Temperatur im Bereich von 275° C bis 25° C.

4. Verfahren nach Anspruch 3, gekennzeichnet durch die Zugabe von Sauerstoff zum pyrolisierten Gegenstand nach dem zweiten Abkühlungsschritt.

5. Verfahren nach Anspruch 3 oder 4, gekennzeichnet durch das Aussetzen des pyrolisierten Gegenstands einer trocknenden Umgebung als letztem Schritt.

6. Verfahren nach einem der Ansprüche 3 bis 5, dadurch gekennzeichnet, daß das pyrolisierte Material mit der Salzlösung durch Vakuumimprägnierung in Berührung gebracht wird.

7. Verfahren nach einem der Ansprüche 1 bis 6, dadurch gekennzeichnet, daß das Cellulosematerial aus Pappe, Papier, Bambus, Eichenblättern und extrudiertem Tabak ausgewählt ist.

8. Rauchartikel mit einer im wesentlichen röhrenförmigen Hitzequelle, die hitzebehandelte Cellulose umfaßt, sowie einem angrenzend an das Mundstück des Gegenstands angeordneten Geschmackserzeuger, der ein wenigstens einen thermisch freisetzbaren Geschmacksstoff enthaltendes Substratmaterial umfaßt, dadurch gekennzeichnet, daß die Hitzequelle eine nach einem Verfahren nach einem der Ansprüche 1 bis 7 hergestellte Kohlenstoffhitzequelle ist, die eine Porosität hat, die ausreicht, die Verbrennung zu unterstützen und eine solche Dichte, daß der zug-induzierte Luftstrom die Röhre passiert.

9. Rauchartikel nach Anspruch 8, dadurch gekennzeichnet, daß das Substrat aus Aluminiumoxid, Tabakfüller, Magnesiumhydroxid, Zeolithen, Glaswolle, Holzkohle, Fuller's Erde, natürlichen Tonen und

aktivierten Tonen ausgewählt ist.

10. Rauchartikel nach Anspruch 8 oder 9, gekennzeichnet durch ein poröses, brennbares Material, das innerhalb der Rohrpassage angeordnet ist und eine Porosität aufweist, die größer ist als die Porosität der Kohlenstoffhitzequelle.