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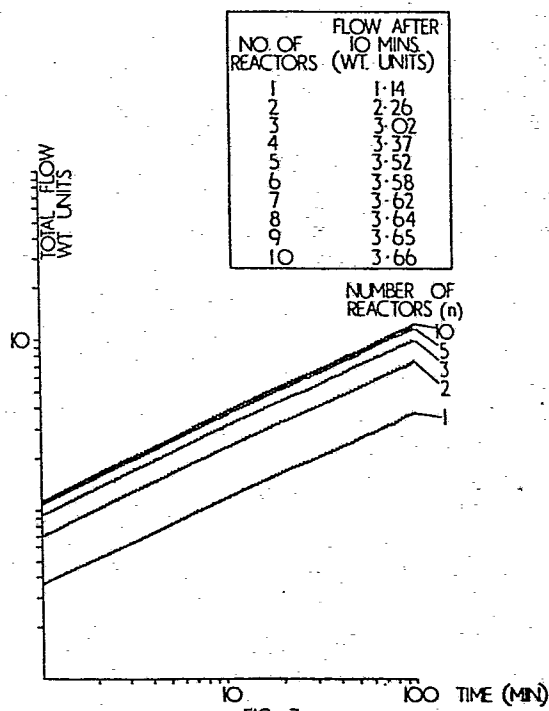
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(54) **Process for liquid extraction of coal.**

(57) Filtration of coal digest can be greatly facilitated by liquid phase extraction using an aromatic solvent oil of coal in at least two stirred reactors in series. Control of residence time is greatly enhanced, reducing or eliminating short residence time digest components which increase filter cake resistivity and long residence time components which have repolymerised and cause loss of yield.

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CASE 4021

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"COAL DIGESTION CONTROL"

This invention is concerned with the liquid extraction of coal, more especially with the control of certain aspects of the extraction conditions in order to facilitate the separation of undissolved particles from the extract and to increase extraction yield.

5 The technique of dissolving coal in liquid extractants, usually a heavy aromatic oil such as a coal tar fraction e.g. anthracene oil, or a highly aromatic petroleum fraction, is well established. Considerable research has been effected on this technique in countries having indigenous coal reserves, especially since awareness of the
10 impending "energy gap" became general. There is no great difficulty in achieving high extraction rates of coal in oil, of the order of 80 to 90% with hydrogenation. Without hydrogenation, such high yields are more difficult to achieve due to the repolymerization of the dissolved coal. For the process to be economically viable it is
15 important to obtain a high extraction yield. It is generally desirable to separate the undissolved matter before further processing, thus lowering the ash content and the sulphur content. The undissolved matter is mainly composed of mineral matter (ash) and certain insoluble carbonaceous substances especially that known as fusain. Considerable
20 difficulty, however, is experienced in this separation and a variety



of methods have been advocated, including filtration, anti-solvent precipitation, centrifuges and gravity settling. The applicants believe that filtration offers the best compromise between processing costs, time and equipment costs.

5 It has previously been suggested in British Patent Specification No. 1,090,556 that the extraction step in a hydrogenative liquid extraction of coal be controlled by considering the relative viscosity of the solution of coal and the solvent itself, and ceasing extraction, and hence transferring to a filtration step, at a minimum value of
10 said relative viscosity. The viscosity of coal extracts or solutions varies considerably with time of extraction. The present applicants, in British Patent Specification No. 1,492,739, disclosed for the first time that the viscosity of coal extract was not necessarily the best criterion for deciding on extraction time, but filtration could be
15 improved by controlling the digestion conditions to ensure that the filter cake had a constant and low specific resistivity. This had in general a more important effect on the ease of filtration than the viscosity of the coal extract.

 The applicants have now confirmed that good control over
20 extraction is extremely important if rapid filtration is to be achieved. It has now been discovered that with a continuous coal extraction process, which would invariably be adopted in large scale commercial practice, there is a very substantial variation in residence time in a coal extraction reactor (digester). This is shown by Fig. 1 of the
25 accompanying drawings which clearly illustrates the distribution of



residence time for a nominal 173 minute residence time, and it is important to note the peak at about 60 minutes. Fig. 1 was the result of tests using radioactive tracers injected into a pilot plant digester feed. It has also been established that there is a strong time
5 dependence factor on filter cake resistivity, the shorter the residence time the greater the filter cake resistivity and the longer the residence time the lower the cake resistivity. Unfortunately, with non-hydrogenative digestion the reduction in cake resistivity is at least partially caused by a polymerisation of the dissolved coal
10 species which reduces the yield. This information is illustrated in Table 1 and Fig. II. Fig. II shows the influence of digestion time on yield and filter cake resistivity, using a hydrogenated anthracene oil, which is a hydrogen donor solvent, and Annesley coal (see below) at a 1:3 coal to solvent ratio and at a temperature of 430°C. The
15 yield as indicated on Fig. 2 is determined by the standard method of quinoline solubility, and shows an almost instantaneous achievement of maximum yield. It is possible to theorise that in the first few minutes of extraction, all the coal which is extractable is converted to a gel-like substance holding within it the insoluble coal and ash
20 particles, and that the gel-like substance is soluble in quinoline but not soluble in the solvent present. As extraction proceeds, the gel-like substance is gradually taken into solution until after approximately 30-60 minutes under the usual conditions essentially all the soluble coal substance is in solution. This theory is supported
25 by experimental work which shows that the yield of filter cake (that

TABLE IInfluence of Extraction Time and Temperature on Cake Resistivity,Batch Process.

Coal: Beynon¹ Coal: Solvent ratio 1:4
 Filtered at 200°C and 138 KNm⁻²

Digestion Conditions		Filtrate visc. at 200°C (cp)	Extraction		Cake Resistivity m Kg x 10 ¹¹
Time (min)	Temp. (°C)		% Gas	% Coal Extracted	
0	-	-	0	12	0.3
15	400	2.1	0	61	4
30		2.4	2	66	4.2
45		1.4	3	67	4.2
60		1.8	2	68	5.3
90		1.3	3		4.2
120		1.6	3	66	2.9
180		-	2	66	1.5
240		1.4	2	66	1.4
360		1.2	7	51	0.2
720		1.1	6	27	0.3
60	430	1.0	2	45	0.04
120	430	1.0	5	36	0.09

1 Prime coking coal from Beynon colliery, National Coal Board, S. Wales Area CRC 301a according to "The Coal Classification System used by the National Coal Board" National Coal Board Scientific Control, London, 1964. 26.2% Volatile matter, 9.8% ash.

* Slurry mixed below 100°C.

is, material not in solution in the solvent) shows a fairly smooth decline curve to close to the minimum at about 60 minutes. Such a theory also goes at least some way to explaining the high cake resistivity of short residence time, as the gel-like particles could
5 deform under the stresses applied in the filter cake and thus inhibit flow.

Reconsidering Fig. 1, the spread of residence time produces some short residence time material with a very high cake resistivity and some longer residence time material with low cake resistivity. In
10 fact, the short residence time component, having high cake resistivity, tends to dominate the filtration process. This is demonstrated by a comparison of filter cake resistivities obtained for the filtration of coal extracts produced from the same coal under similar conditions but from difference process systems, i.e. continuous flow extraction
15 and batch extraction. A batch extract of Beynon coal (400°C, 60 min. residence time) had a cake resistivity of $53 \times 10^{10} \text{ m Kg}^{-1}$ and at 430°C the resistivity had fallen to $0.4 \times 10^{10} \text{ m Kg}^{-1}$ (see Table I). However, using the same coal, oil etc. except using a continuous digester for a nominal 60 min. residence time at 410 to 420°C gave
20 a filter cake resistivity under identical filtration conditions of $60 \times 10^{10} \text{ m Kg}^{-1}$. The difference is even more marked in a further experiment using a high volatile coal from Annesley colliery (CRC 702, 34% volatile matter, 3% ash), which gave cake resistivities of 20×10^{10} and $90 \times 10^{10} \text{ m Kg}^{-1}$ for the batch and continuous systems
25 under otherwise identical digestion and filtration conditions.

The present inventors concluded that the spread of residence times is a crucial factor in achieving low filter cake resistivity and hence rapid filtration. This spread would be avoided if the reactor used for extraction gave plug flow, but this would be
5 essentially impossible to achieve on the large scale at which commercial coal extract plants would operate.

This invention provides a process for the continuous production of coal extracts which comprises digesting a coal using an aromatic solvent oil under conditions at which the solvent oil is liquid and
10 thereafter filtering from the resulting digest substantially all the undissolved material to yield a filtrate of extracted coal substance in solvent, the digestion being carried out on a continuous basis in at least two stirred reactors in series, whereby the resistivity of the filter cake is minimised. Preferably, from 3 to 10 stirred
15 reactors in series are used, more preferably 4 to 7, especially 5, stirred reactors in series are used. Conveniently, all the reactors are of equal size and the same conditions obtain in each but this need not be so, and individual designs may use different sizes and/or conditions for some, or all, of the reactors.

20 The use of a plurality of stirred reactors in series amounts to a close approximation to plug flow in a single reactor. The actual number of reactors will depend upon economic aspects, especially on the materials being processed and capital costs. It is economically viable to increase the number of reactors until the extra extraction
25 costs become equal to the cost saving in filtration, that is

considering each reactor relative to the previous reactor so that the saving in filtration cost > increased digester costs.

The choice of reactors and the method of stirring is a matter for normal chemical engineering design. It may be convenient, for example, to construct theoretical reactors within a single reactor shell, thereby minimising costs allied to the maintenance of elevated pressure and temperature.

In theory, a turbulent flow reactor should provide plug flow and sufficient mixing, but this is not achieved in practice, so that stirring is required. It is important that adequate mixing occurs in the digestion, if this is insufficient, a much slower filtration will result. This is illustrated in Table II below, in which identical reaction conditions were used, in the same digester, with and without stirring.

Table II

Digest	Stirring	% Extraction Yield	Cake Resistivity (m Kg ⁻¹ x 10 ¹⁰)
Beynon coal/anthracene oil	Yes	68	53
	No	60	194
Annesley coal/hydrogenated anthracene oil	Yes	88	26
	No	84	423

In the process of the present invention, the solvent oil is preferably a coal tar fraction or a highly aromatic petroleum fraction or a hydrogen donor solvent, and these classes of solvent are well known in the art. The coal is suitably a high to low rank bituminous coal, but the invention may be applied to other coals such as brown

coal. Reaction conditions may be selected within those known in the art after consideration of filter cake resistivity and extraction yield.

It will be understood that the process of the invention may
5 take place in the presence or absence of free gaseous hydrogen. It is believed, although this invention is not to be limited by the theories expressed herein, that the initial and primary action of hydrogen is to hydrogenate the solvent oil, which then transfers the hydrogen to the coal substance. That is, the solvent acts as a
10 hydrogen donor. It is therefore believed that the experimental work which formed the basis of the present invention illustrates the applicability of the method to both hydrogenative and non-hydrogenative extraction of coal.

In general, in the temperature range of 350 to 450°C, higher
15 temperatures require lower residence times.

To illustrate the invention, the prime coking Beynon coal is extracted in anthracene oil at 430°C for a 20 minute residence time, then filtered at 200°C and 138 KNm⁻².

The filtration rate for a number of extractions, using a varying
20 number of identical reactors in series, with identical conditions otherwise, has been plotted as total flow against time, in Fig. 3, using log scales. It will be observed that there is a substantial improvement in using two reactors compared to a single reactor, also that little additional improvement can be expected beyond 10 reactors.

25 With a non-hydrogenated system the extraction yield is dependent

on the time of digestion. A longer digestion time results in the polymerization of the dissolved coal and a reduced yield. Therefore, a spread of residence time will reduce the extraction yield since repolymerised material is formed. By using several stirred reactors, almost plug flow can be achieved and an increased extraction yield. Extraction yield is of considerable importance in the overall economics of a coal digestion process, and even small improvements are worthwhile. Repolymerisation of coal moieties is relatively slow with high rank coals (cf Table I) or when using a hydrogen donor, but is relatively fast when digesting low rank coals. Thus, especially with low rank coals, the use of several stirred reactors according to the invention has the additional benefit of increased extraction yield.

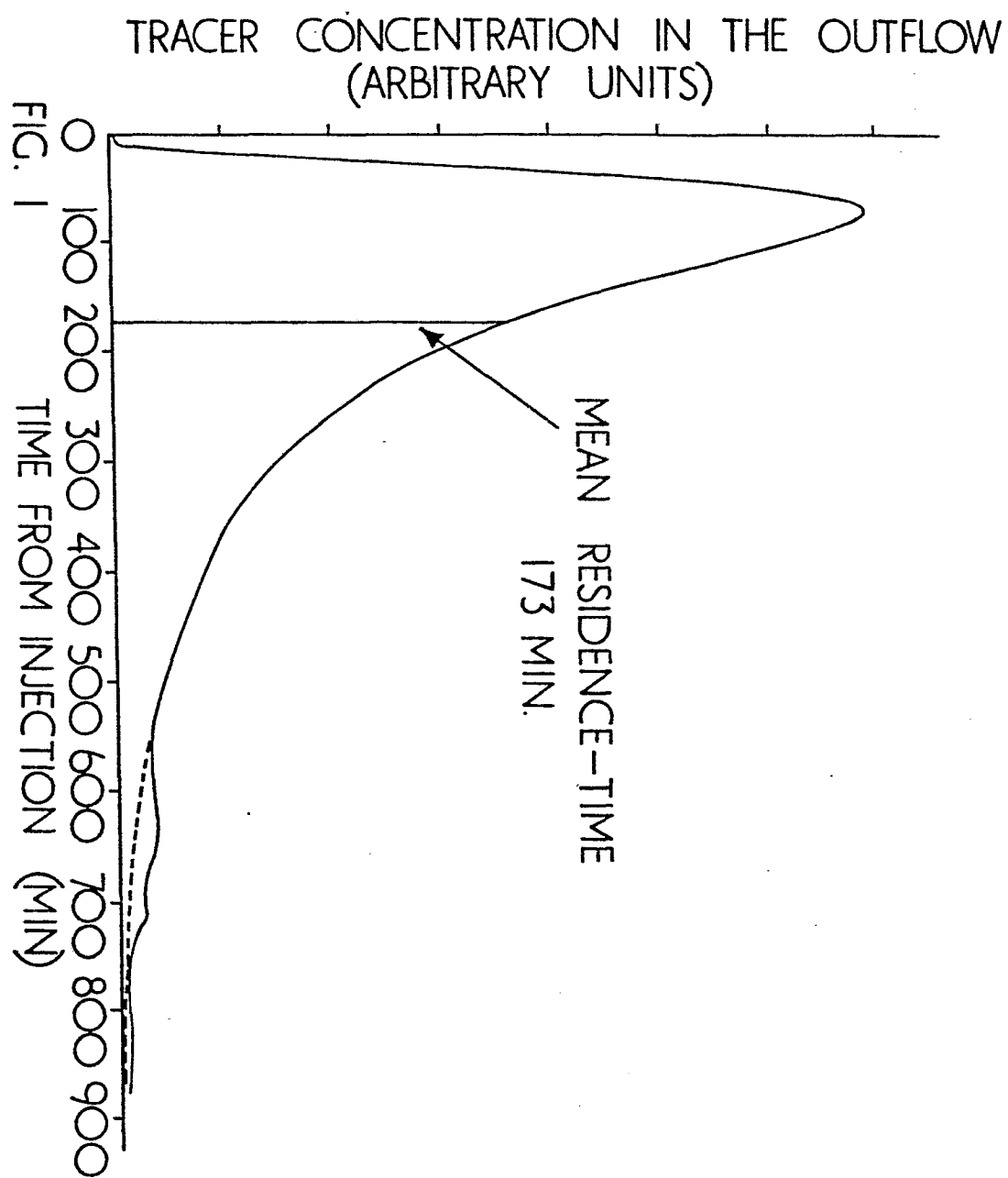
It is therefore shown that the mere increase in the number of reactors according to the invention can lead to a substantial and unexpected increase in the rate of filtration; the overall throughput of a coal extraction plant can be increased or the filter reduced in size. An increase in extraction yield flows as a further benefit from the invention.

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PATENT CLAIMS:

1. A process for the continuous production of coal extracts by digesting on a continuous basis a coal using an aromatic solvent oil under conditions at which the solvent oil is liquid and thereafter filtering from the resulting digest substantially all the undissolved
5 material to yield a filtrate of extracted coal substance in solvent, characterised in that at least two stirred reactors in series are used, whereby the resistivity of the filter cake is minimised.
2. A process according to claim 1, characterised in that from 3 to
10 stirred reactors in series are used.
3. A process according to claim 2, characterised in that from 4 to
7 stirred reactors in series are used.
4. A process according to any one of claims 1 to 3, characterised in that the digestion is carried out at a temperature within the range of 350 to 450°C.
- 15 5. A process according to any one of claims 1 to 4, characterised in that the solvent oil is a coal tar fraction, a highly aromatic petroleum fraction or a hydrogen donor solvent oil.
6. A process according to claim 5, characterised in that the solvent oil is anthracene oil or a hydrogenated anthracene oil.



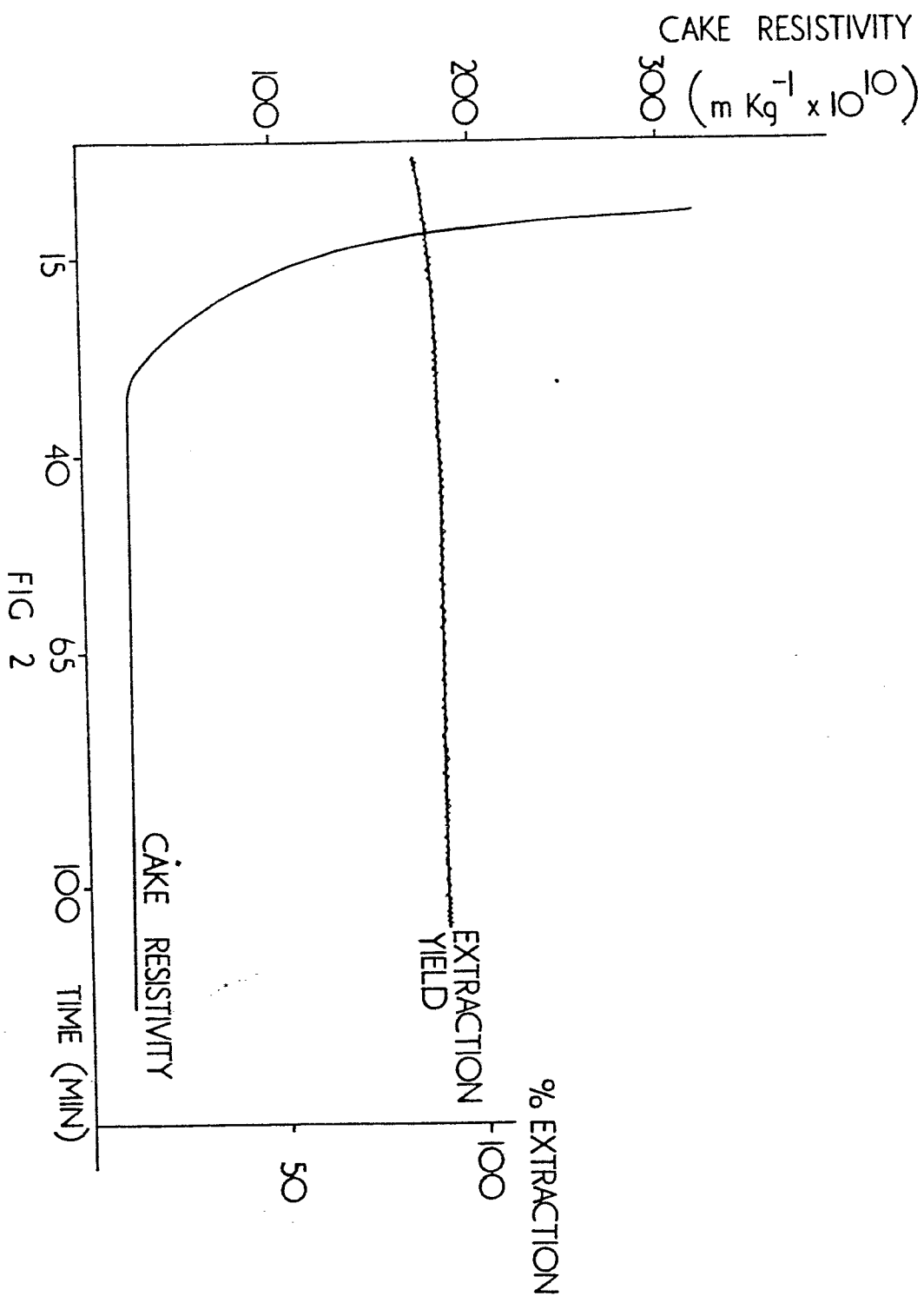
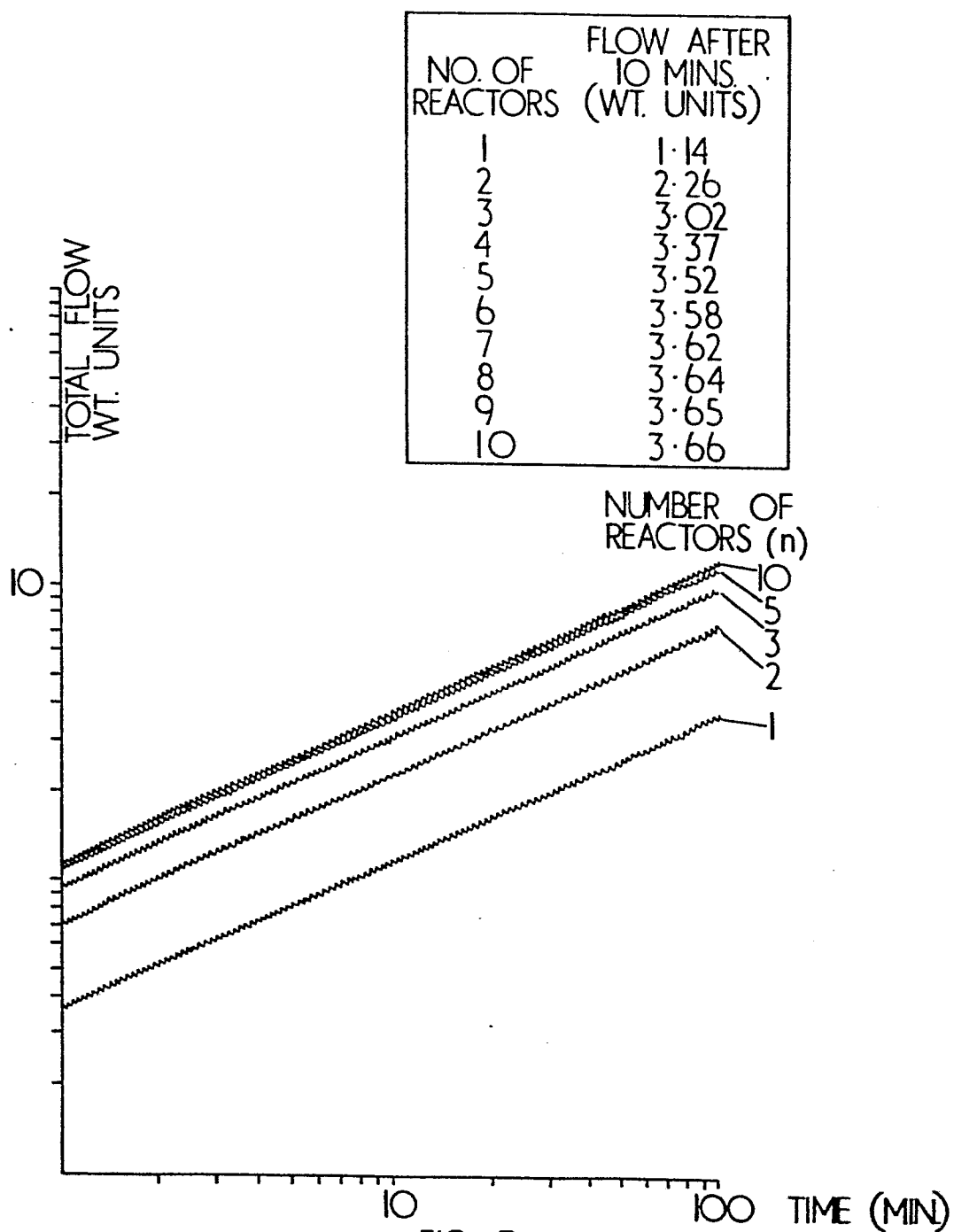


FIG 2





European Patent
Office

EUROPEAN SEARCH REPORT

Application number
EP 79 30 2933

0014804

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int. Cl. 3)
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
	<u>DE - C - 705 178</u> (I.G. FARBEN) * Page 1, line 45 - page 2, line 14; claim *	1,4,5	C 10 G 1/04
	-- <u>US - A - 3 594 304</u> (SEITZER et al.) * Claims 1-3 *	1,4-6	

			TECHNICAL FIELDS SEARCHED (Int.Cl. 3)
			C 10 G 1/04 1/00
			CATEGORY OF CITED DOCUMENTS
			X: particularly relevant A: technological background O: non-written disclosure P: intermediate document T: theory or principle underlying the invention E: conflicting application D: document cited in the application L: citation for other reasons
			&: member of the same patent family, corresponding document
b The present search report has been drawn up for all claims			
Place of search The Hague		Date of completion of the search 10-04-1980	Examiner DE HERDT

